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CHOLNOKY JENŐ KARSZT- ÉS BARLANGKUTATÁSI PÁLYÁZAT
Bíráló Bizottsága

Debrecen, 1998. február 4.

Tisztelt Bíráló Bizottság,

Az 1998-ban meghirdetett ***CHOLNOKY JENŐ KARSZT- ÉS BARLANGKUTATÁSI PÁLYÁZATRA*** **egyéni kategóriában** az 1997-es munkásságommal, két megjelent cikkel, egy könyvfejezettel, PhD disszertációmmal valamint 1997 őszén egy nemzetközi konferencián elhangzott előadásommal pályázok. A pályázati anyagom egyes tételeit, elősegítendő az áttekintést, röviden a következőképpen jellemezhetem.

1. Hakl J. *et al.* Radon Transport in Fractured Porous Media - Experimental Study in Caves. *Environment Intarnational* **22** (1996) S433-S437
A nyomdai késések miatt ez az anyag a feltüntett évvel ellentétben csak 1997 márciusában jelent meg. Emiatt tettem bele a pályázati anyagba. Maga a cikk a külső átlagos napi hőmérséklet függvényében kapott napi átlagos barlangi radonkoncentráció görbékket hasonlítja össze az Abaligeti-, Vass Imre- és Szemlő-hegyi-barlangokra. Alapvető kapcsolatot állapít meg barlangok szerkezete és ezen görbék lefutása között, melyek tükrözik a külső környezeti hatások felszín alatti terjedését. A cikk kitér a Cserszegtomaji kútbarlangban kapott gyors időbeli változások okára.

2. Hakl J. *et al.* Radon Monitoring in Caves. Fejezet a "Radon Mesurements by Etched Track Detectors, Applications in Radiation Protection, Earth Sciences and the Environment" c. könyvben. Szerk.: S.A.Durrani és R.Ilic, World Scientific, Singapore, 1997, 261-283
A fejezet a szerkesztők felkérésére készült a barlangi radon irodalom áttekintése céljából. Ezzel együtt a fejezet 95%-ban a saját, magyarországi barlangokban nyert tapasztalatainkra és eredményeinkre épít. A fejezet elején áttekintem az irodalomban fellelhető barlangi radonmérések eredményeit, hozzávéve a saját, még nem publikált eredményeinket, ismertetem a Hajnóczy-, Létrási-Vizes, Cserszegtomaji-kút- és Sátorkő-puszta-barlangban megfigyelt

hosszúidejú változásokat, vizsgáltam a radon nyomjelzőkénti használhatóságát barlangi környezetben, a detektált transzport folyamatok környezeti- és környezetvédelmi aspektusait, a gyorsidejű változásokat, azok terjedését a Vass Imre-, Cserszegtomaji-kút és Abaligeti-barlangokban. A fejezet érinti az Abaligeti-, Béke-, Szemlő-hegyi, Szent István-, Tapolca- és Baradla-barlangokba tapasztalt radonszintek sugáregészségügyi következményeit és 61 tételes irodalomjegyzékkel zárul.

3. Hakl J. *et al.* Radon Transport Phenomena Studied in Karst Caves - International Experiences on Radon Levels and Exposures. *Radiation Measurements 28* (1997) 675-684
A cikk ez előbbi könyvfejezet rövidített és két helyen bővített változata. A két bővítés a Szemlő-hegyi-barlang radon idősora és a felszíni hőmérséklet közötti párhuzamot taglalja, valamint részletesen ismerteti a Vass Imre-barlangban a szifon záródásakor megfigyelt radonszintek és a barlang szerkezete közötti kapcsolatot.

4. Hakl J. Application of Radon-222, as a Natural Tracer in Environmental Studies. PhD értekezés. Debrecen, 1977, 30 oldal. A dolgozatban tézisszerűen összefoglaltam a radontranszport vizsgálatok során nyert eddigi eredményeimet. A hat tézispont közül öt (összesen 11 alpont) a barlangi radonmérések során nyert eredményeimre vonatkozik.

5. Hakl J. *et al.* Site Specific Radon Regimes of Cave Systems. Szóbeli előadás a "Rare Gas Geochemistry" c. IV nemzetközi konferencián (1997. október 8-10, University of Roma TRE). Az előadás anyagát mellékeltem. Az előadásban a barlangi radon idősorok analízise alapján vizsgáltam a barlangi mikroklimatikus zónákat, azok stabilitását, a külső hatások behatolását a felszín alatti környezetbe; a barlangjáratok morfológiája és a külső hatások terjedése közötti kapcsolatot. Név szerint érintettem a Vass Imre-, Baradla-, Pálvölgyi-, Mátyáshegyi-, Sátorkő-puszta-, Létrási-Vizes, Cseszegtomaji-kút- és Alba-Regia-barlangokat.

Tisztelettel:

Hakl József

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RADON TRANSPORT IN FRACTURED POROUS MEDIA - EXPERIMENTAL STUDY IN CAVES

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The spatial and temporal variation of ^{222}Rn concentration in three horizontal caves and in one vertical cave was measured to study the influence of different morphological and meteorological parameters on the forming of airborne radon concentrations inside. In horizontal caves, the daily mean radon concentration as a function of the daily average surface temperature showed a step-function type dependence with low winter and high summer values reflecting the main direction of underground airflows. Restriction of airflows increased winter but decreased summer radon levels. The transition pattern between the low winter and high summer values gradually linearized as the number of vertical fractures communicating with the surface increased. Contrary to horizontal caves, in the vertical cave barometric pressure variations played the most important role in controlling subsurface radon concentrations. Decreasing pressure increased radon levels, and increased pressure decreased radon levels. In the pressure-radon correlation curve, there was a small hysteresis which indicated the nonlinearity of the process.

INTRODUCTION

Karstic strata have a characteristic feature: due to the infiltrating waters, a special chemical dissolution process takes place in the rock matrix. The phenomenon results in the formation of an interconnected set of larger openings and fractures. A cave is a part of this system (Ford and Williams 1989). Due to the special morphology formed, the cave is able to communicate through the overburden under the influences of changing atmospheric pressure or temperature differences between cave and surface. The morphology- and meteorology-linked flows play a significant role in governing the underground radon transport in these areas. The flows are practically restricted to the fractures, as 99% of the flow is carried by the cracks embedded in the rock matrix (Nilson et al. 1991). From the standpoint of morphology, two basic situations were considered, depending on the main physical process driving the flow:

Type A: Horizontal caves connected to the surface with one or more fractures, shafts. In this case, the pressure exerted at the entrance by the column of air inside the cave will differ from the pressure of external air because the density of the air depends upon temperature. The pressure difference in the first approximation is proportional to the temperature difference between the cave and outside. Airflow is controlled by the chimney effect.

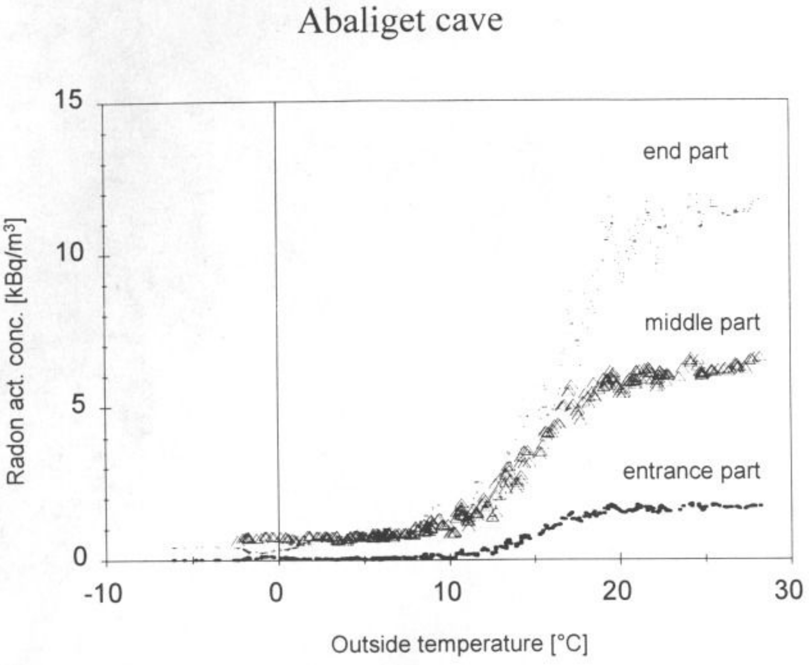

Fig. 1. Daily mean radon concentrations in the Abaliget cave as a function of daily mean surface temperature. The width of transition is approximately 10°C.

Type B: Vertical cave. In this case, the passageways lie below the entrance and atmospheric pressure is the main driving parameter for the flows. The volume of cave air breathed out (in) is proportional to the difference in atmospheric pressure changes and cave volume.

For horizontal caves, the volume of daily breathed in/out cave air can be comparable to the cave volume (Fodor 1981). In vertical caves it can be on the order of a few percent (1-5%) of the cave volume. This suggests that advection is the dominating physical process governing the radon transport, which in turn may be affected by the size and density of openings and fractures (Nilson et al. 1991; Schery and Siegel 1986).

MATERIALS AND METHODS

For the present work, a microprocessor-controlled automatic multichannel field radon monitor (Dataqua Systems, Dataqua Ltd., Hungary, Kölcsey F. u. 1, 8220 Balatonalmádi) was used with simultaneous registration of cave temperature and air pressure. Radon was measured with an open type diffusion chamber (delay time ≈ 1000 s) equipped with alpha sensitive Si based semiconductor detectors. The sensitivity of the unit was one count per hour (cph) at 54 Bqm^{-3} of radon with an 0.1 cph initial background. Approximately 200 000 hourly readings were obtained with 11 monitors in the four caves investigated during the years 1992-94. The studied caves were:

1) Vass Imre and Abaliget caves are two cases similar to the model cave of type A. They have one entrance and the main passage is situated horizontally. In the middle of the Vass Imre cave, there is a siphon, which was closed by water two times during the observation period. Three radon monitors were operated in each cave: one at the entrance, one in the middle, and one at the end of the main passages.

2) The Szemlő-hegy cave is a horizontal cave with entrances at the foot of the hill and at the top of the hill. The cave passageways are situated in two levels and the cave can be considered as a cave of type A, with one dominating large opening to the surface. One monitor was operated at each level of the cave.

3) The Cserszegtomaj well cave is a horizontal labyrinthine cave which was formed at 50 m depth on the boundary of dolomite and sandstone. It has one 50 m deep artificial vertical entrance (type B). The whole formation is covered by clay. The radon concentration is unusually high due to the sandstone environment of high porosity and bad ventilation. Five radon monitors were operated in the cave: one at the bottom of the entrance, and the other four in the depth of the cave.

Additional site specific surface meteorological data were obtained from the Hungarian National Meteorological Service.

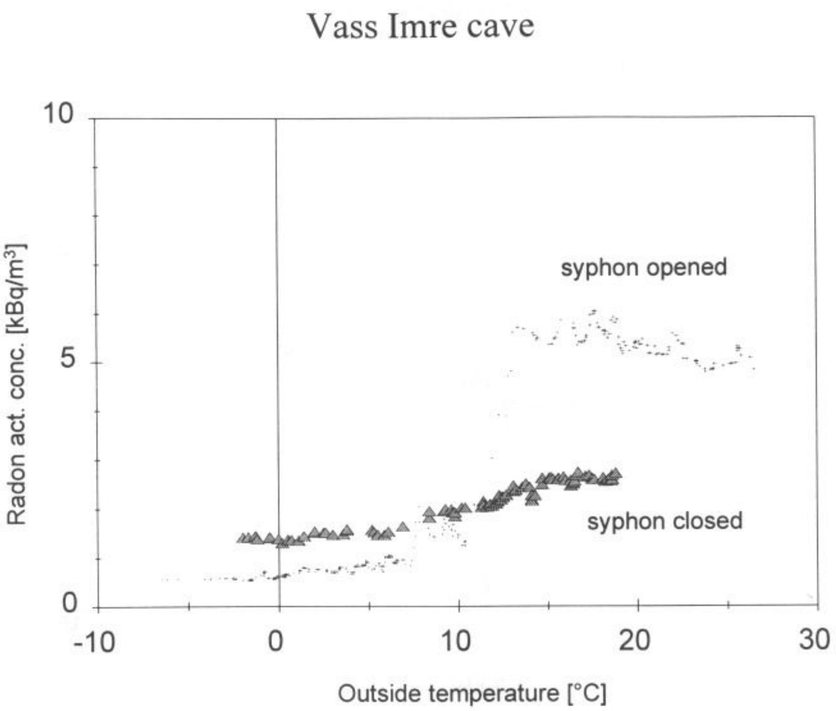

Fig. 2. Daily mean radon concentrations in front of the siphon in the Vass Imre cave. The width of transition is approximately 5 °C.

RESULTS AND DISCUSSION

In the horizontal caves, there is continuous airflow through the entrance either in or out (in winter and summer, respectively). A typical dependence of daily mean radon concentration on the daily mean outside temperature is shown in Fig. 1.

The daily mean radon concentration follows an S-type dependence on the outside temperature. The radon levels increase with the distance from the cave entrance. The observed pattern unambiguously reflects the change in the main direction of underground airflows.

The same type of behavior can be found in the Vass Imre cave. In this case, however, the volume of infiltrated air is controlled also by the penetrability of the siphon. This phenomenon, in turn, markedly affects the radon levels, as seen in Fig. 2.

When the siphon is closed, there is practically no airflow through the entrance. The restriction of airflows has a different effect on the radon level depending on the season. In summertime, it decreases and, in wintertime, it increases the daily mean radon levels. More remarkably, the airflow restriction also result in an overall drop of 30% in the annual mean radon concentration. This asymmetric effect can be explained on the basis of the air circulation model (Géczy et al. 1988) and is in agreement with the results of numerical calculations (Holford et al. 1993).

The spatial distribution of radon concentrations along the main passage of the Vass Imre cave is uniform, contrary to the Abaliget cave. The difference between the two caves can be attributed to the differences in the number of vertical fractures communicating to the surface. In the Vass Imre cave, only one less developed fracture system, located towards the end of the cave after the siphon, is known (Holly et al. 1956). In the Abaliget cave, more than one better-developed vertical fracture system exists (Szabó 1961).

The number of fracture systems also has an effect on the widening of the transition phase. While in the Vass Imre cave, this width is around 5 °C, in the Abaliget cave, it is around 10 °C. This step-by-step widening of the transition state depending on the number and size of openings is further supported by the data obtained in the Szemlő-hegy cave (Fig. 3). Here, the S-type curve is practically reshaped to a line. However, the turn in the radon curve measured at the upper situated passage indicates the proximity of the surface. From the point of view of radon transport, the equilibrium (saturation) value is not yet reached at this level of the cave.

In vertical caves, changes in surface temperature have only a small effect on radon concentration inside the cave and only on an annual scale. Atmospheric pressure has a strong influence on the radon level, and direct pressure changes play an important role in controlling radon trans-

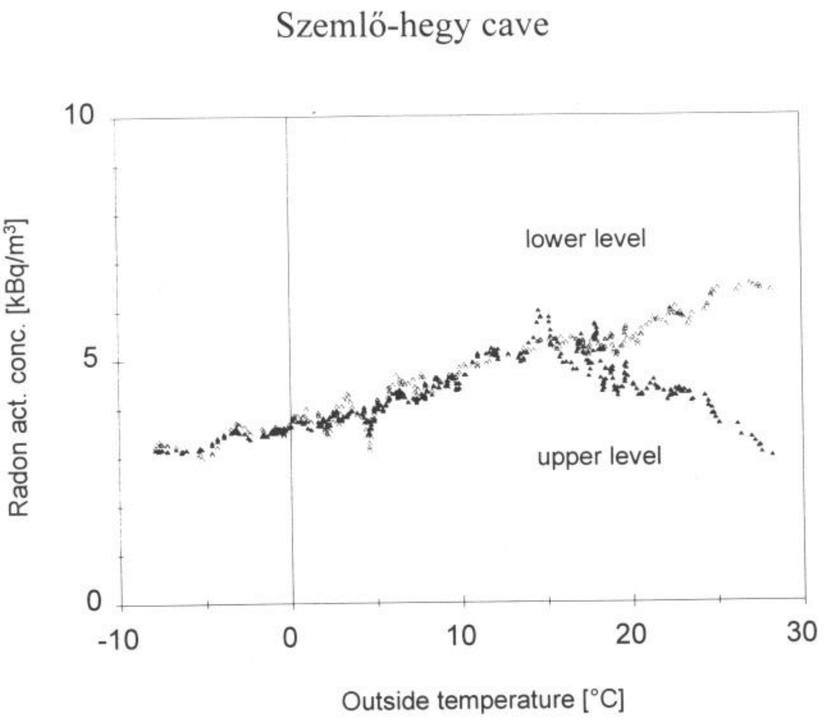

Fig. 3. Daily mean radon concentrations in the Szemlő-hegy cave.

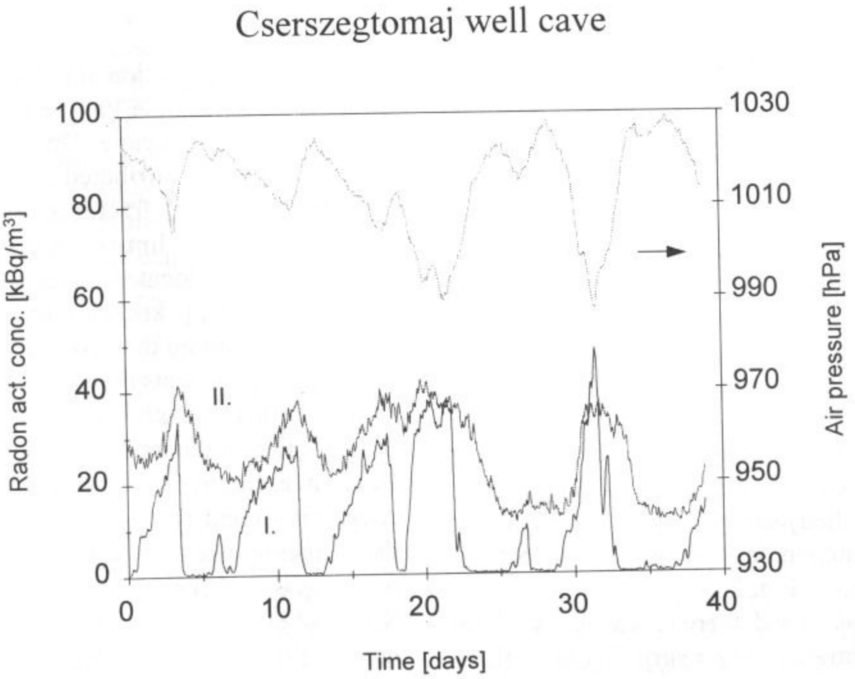

Fig. 4. Atmospheric pressure and radon time series (hourly readings) in the Cserszegtomaj well cave. Site I is located 50 m below the surface near the artificial entrance; Site II is at the same level but at the cave end.

port. The radon time series, observed in the Cserszegtomaj well cave, markedly shows this effect (Fig. 4). Radon transport is predominantly controlled by advection, as shown by the immediate response of radon concentrations to pressure changes. Decreasing pressure increases the radon level and inversely. The response is almost immediate. The small hysteresis found in the pressure-radon correlation curve indicates the nonlinearity of the processes. It corresponds to a time shift in the response of airflow velocity to the pressure changes (Wigley et al. 1967).

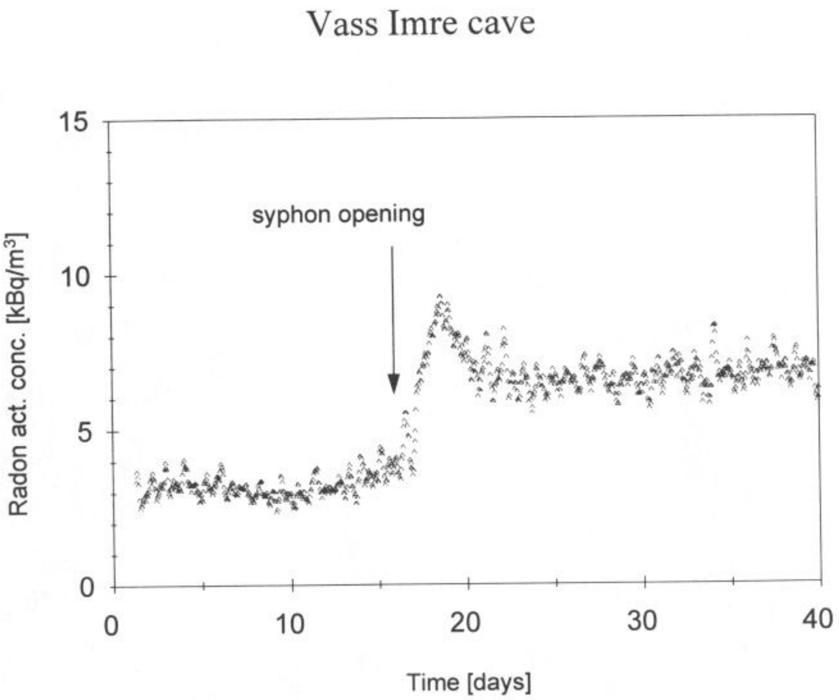

Fig. 5. Time series of hourly mean radon concentrations in front of the siphon in the Vass Imre cave with the effect of opening the siphon.

Analyzing temporal changes in the radon time series, advection was found to be the dominating transport process. This is also shown on the transient part of the radon record from the Vass Imre cave, which corresponds to a summertime siphon opening (Fig. 5). The shape of the curve, compared to results of numerical calculations on radon exhalation from porous media (Janssens et al. 1984), corresponds to the case when a sudden pressure drop results in instantaneous airflow development. The latter effect was also experimentally observed during an artificial siphon closing and opening.

CONCLUSIONS

Field measurements of real-time radon monitoring in caves showed that, in karstic regions, radon carried by subsurface fluids may migrate over longer distances along caverns and fractures. The possible transport lengths depend on the velocity of carrier fluid as well as on the geometrical size of the openings. The advection is driven by factors of meteorological origin. Temperature differences formed between cave and outside air induce air circulation in horizontal caves resulting in temporal variations in the radon records. Atmospheric pressure changes have a more significant effect on the radon content of vertical caves.

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RADON MEASUREMENTS
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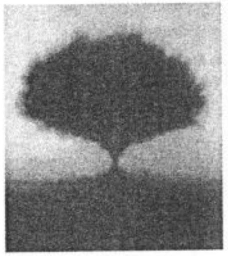
Applications in Radiation Protection, Earth Sciences and the Environment



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*To all those who have contributed to a better understanding of radon,
its effects, and its applications.*

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4.1 Radon Monitoring in Caves

4.1.1 Introduction

Caves are commonly considered as static environment, where not only the continual darkness but the temperature and high relative humidity are stable. In most cases this belief is not justified, because air currents can cause measurable changes in the physical parameters. Caves occur everywhere on the earth, although mainly in karst areas - as they are formed mostly in limestone environments. It is obvious that the noble gas radon, which moves freely through the pores of permeable rocks, will easily penetrate into subsurface cavities and even huge caves.

In the last two decades the development and the widespread application of etched track techniques have found their place also in radon measurement projects aimed to study the cave environment. These measurements have ranged from sporadic observations up to regular long-term studies, and they were motivated by dosimetric and transport study approaches as well. In Hungary the first *in situ* field radon measurements in caves performed by etched track techniques started as early as 1977 and were initiated by Dr G. Somogyi, one of the pioneers on the etched track field. Since then, the number of studied caves has increased substantially and we have reached 10,000 observation data in 31 Hungarian caves.

After recognising the link between the secondary porosity, fracturization and formed radon levels the ever-pressing interest to understand underground radon transport in karstic environment has been increased. During the last several years active electronic devices, automatic multiparameter (radon - typically one hour integration time, pressure, temperature, humidity) monitors (Dataqua Ltd, Hungary, Kölcsey F. u. 1, 8220 Balatonalmádi), have been included into cave studies. These studies were partly financed by Hungarian funds (projects AKA 1-3-86-185, OTKA 2011, 3005 and T 016558, T 017560) during the course of the last ten years. The accomplishment of this section thanks is to this support, where of course international experiences are also appreciated.

4.1.2 Sources of Radon Isotopes in Cave Environment

The sources of radon in caves are the bedrock and deposits. Radon levels in caves are influenced primarily by the uranium content of the rock. Limestone and other sedimentary rocks are found to contain about 1.3-2.5 ppm ^{238}U (16-31 Bq kg^{-1}) on average.

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The relatively high values of radon found in caves are due to these minute quantities of parent substances that occur naturally on and within the interior surfaces of the caves. Increased ^{238}U concentrations can be associated either with fluorite mineralizations or hydrocarbons present in the surrounding limestone.

Uranium can be oxidised and mobilised by groundwater flow. Once reducing conditions are encountered, the uranium is readily precipitated from solution. This leaching fixation process leads to the enrichment of uranium in adjacent deposits. This secondary transport and enrichment process is important in cave environment, as fractures in rock can increase the surface area interacting with water. Experimental evidence of this effect in cave environment was found by Navrátil *et al.* (1993), who observed enrichment of uranium on cave walls.

The primary radon source is the ^{226}Ra content of the rock. Nazaroff *et al.* (1988) report mean ^{226}Ra content of carbonates to be 25 Bq kg^{-1} (range 0.4-233), which were found to be distributed lognormally. The reports of other authors (Wilkening, 1981): 96.2 Bq kg^{-1}; (Surbeck, 1990): 20 Bq kg^{-1}, well agree with this value. The results of ^{226}Ra determinations of bedrock and soil samples from the Hungarian caves examined fall in range 0.6-26 Bq kg^{-1} (Dezső, 1995). The relatively impermeable soils (deposits) such as clay do not have sufficient porosity to allow transfer of significant amounts of soil gas; therefore their contribution to radon budget is small (Michel, 1987). Accordingly, Burkett (1993) found radon emitted from the clay to be not sufficient to account for the radon concentration measured in the cave.

4.1.3 Review of Some Published Radon Data in Caves

Continuous radon measurements covering at least one-year long period were performed by the etched track technique in Hungary, Italy, Slovakia and Luxembourg. Seasonally one-week-long observations were performed in England, and measurements not covering a full year are reported from Mexico and the USA.

Based on long-term continuous radon observation the relative variations of radon between monitor locations indicate that the cave atmosphere is not a uniform radon environment over any given time period; so it is very hard to represent one cave with one radon value. Therefore, we decided to compile all the globally available radon data from 220 different caves (Ahlstrand, 1980; Amano *et al.*, 1985; Borau *et al.*, 1993; Burkett, 1993; Cappa *et al.*, 1995; Cigna and Clemente, 1981; Collar and Odgen, 1991; Cunningham and LaRock, 1991; Fernandez *et al.*, 1984; Géczy *et al.*, 1988; Gunn *et al.*, 1991; Hakl *et al.*, 1992; Hakl, 1993, 1994; Hyland and Gunn, 1994; Hyland *et al.*, 1994; Kobal *et al.*, 1986, 1987, 1988; Lénárt *et al.*, 1988; Kaufmann *et al.*, 1995; Lively and Krafthefer, 1995; Marx, 1996; Massen *et al.*, 1995; Miki and Ikeya, 1980; Navrátil *et al.*, 1994; Papastefanou *et al.*, 1986; Quinn, 1988, 1990; Roda *et al.*, 1994; Seymore *et al.*, 1980; Solomon *et al.*, 1992; Surbeck, 1990; Vibranov *et al.*, 1975; Vičanová *et al.*, 1994; Wilkening, 1979, 1980; Wilkening and Watkins, 1976; Yarborough, 1980, 1981) including our not yet published data from 12 Hungarian caves. In the distribution represented in Fig. 4.1.1, we have included either cave minimum and maximum data (given by authors) or average values. The distribution of the data is close to lognormal.

We note that, taking into account results obtained only by etched track methods, the "annual average" radon concentrations in karstic caves range from 0.1 to 20 kBq m^{-3} with a 2.8 kBq m^{-3} arithmetic average. The lower end of the scale is associated with big chamber volumes or high ventilation rates; the upper end is characterised by closed, badly ventilated places and uranium-rich sediments. The highest annual average was observed in 1992 in the Cserszegtomaj well-cave, Keszthely Mountains, Hungary. The maximum two-monthly average reached 33 kBq m^{-3} in the same cave. On the other hand, Hyland and Gunn (1994) report 46 kBq m^{-3} one-week average from the Peak District region, England. The maximum radon concentration is also reported from the same region, viz. from Giants Hole, Derbyshire Peak District, by Gunn *et al.* (1991). The result of the spot measurement was 155 kBq m^{-3}.

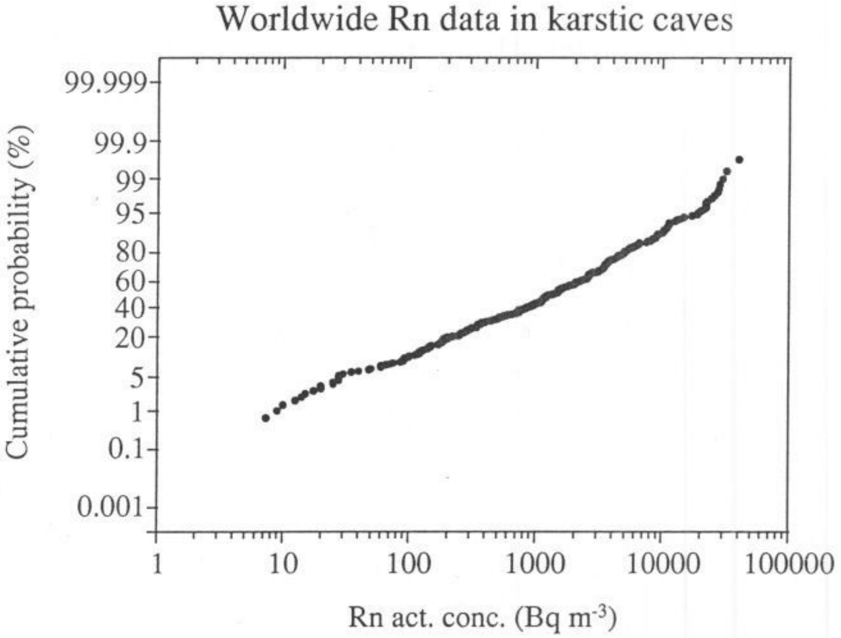

Figure 4.1.1: The distribution of radon activity concentrations reported from different caves worldwide.

4.1.4 Techniques of Radon Measurement in Caves

For the purpose of cave radon measurement, different types of opened and closed diffusion chambers are used, equipped with alpha sensitive polymer track detectors. Somogyi *et* *al.* (1983) described different types of multi-detector devices suitable for measurement in caves. The most significant improvement in the measuring technique was the thermally stabilised double-wall diffusion chamber (filled with water), which avoided the frequent problem of wetting of detector surfaces. Nowadays several commercially available radon monitors developed for indoor radon measurements are used in cave environment, which, owing to the high relative humidity, may not always be optimal. The typical exposure time ranges from one week to several months.

From the beginning of the 1990's, diffusion chambers equipped with Si based semiconductor detectors and connected to microprocessor controlled data storing units were also introduced into cave studies. Their time resolution is limited by diffusion and detection efficiency; typically it is one hour.

4.1.5 Some Experimental Results and General Trends for Long-Term Monitoring

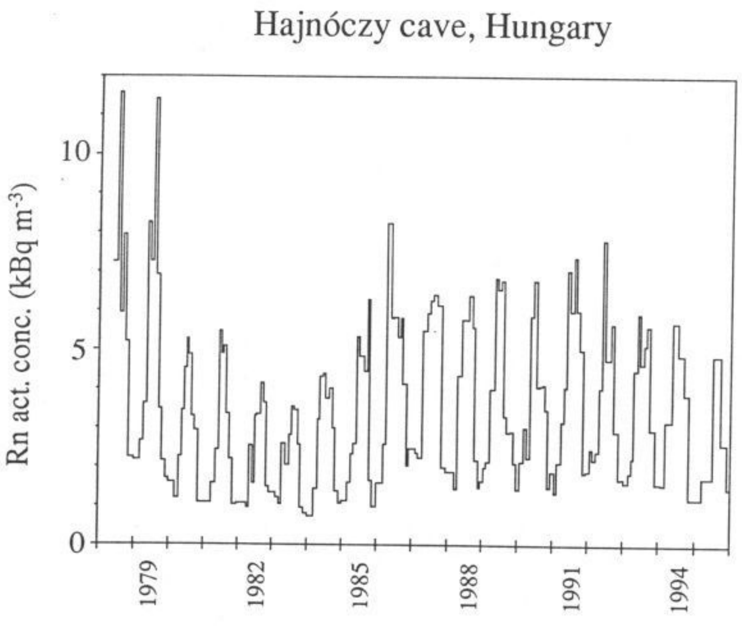

Figure 4.1.2: Radon time series observed by using etched track technique in the Great Hall of the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary. Each observed value is the average of three simultaneous readings with one month integration time.

It is a general phenomenon that in each cave more or less periodical fluctuations of smaller or larger amplitude can be found around the mean value of the radon activity concentration. The frequency and amplitude distributions of the data are characteristic of the cave and its environment, of the uranium (radium) content of the enclosing rocks and stones and of the extension of that porous surrounding which is in connection with the cave air by the intrusion of atmospheric air and radon traced subsurface fluids.

The longest time series were obtained by us in the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hunary (Fig. 4.1.2), which shows a very regular seasonal variation. It is clearly seen in the figure that the seasonal variation is superposed on a long-term change of the mean activity concentration. The same effect was found in a few other Hungarian caves; but it is also reported by Lively and Krafthefer (1995). In Fig. 4.1.3, the annual average radon data from three different types of caves are shown.

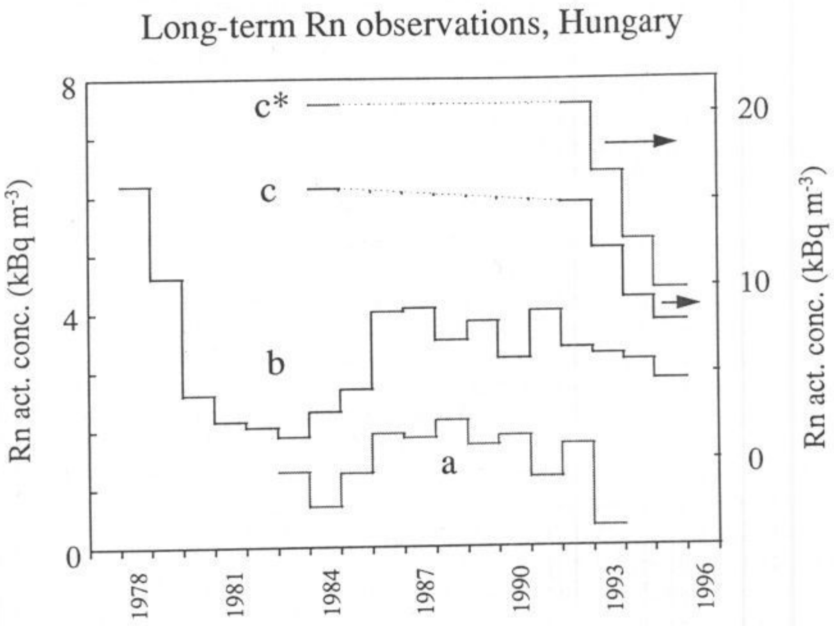

Figure 4.1.3: Long-term change observed in three Hungarian caves of different types. Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hunary, is dominated by chimney effect winds; Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary, is characterised by temperature difference driven convective air motions; and Cserszegtomaj well-cave, Keszthely Mountains, SW Hungary, is influenced mainly by atmospheric pressure changes. The curves represent the middle part of the Létrási-Vizes cave (a); Great Hall in the Hajnóczy cave (b); and cave average (c) and a remote part (c*) in the Cserszegtomaj well-cave.

The long-term change can be amplified selectively at different test sites of a given cave. This effect is markedly shown on time series taken at two different depths of a vertical cave (Sátorkő-puszta cave, Pilis Mountains, N Hungary). The relative difference of the two data series [(a-b)/((a+b)/2) x 100%] shows an increasing tendency with years (see Fig. 4.1.4).

The observed phenomenon displays the effect of slowly changing environmental parameters on radon transport processes. Such an external parameter may be the change in radon emanation power due to the slow change of water content of the cave surrounding, which may reflect the variation in annual precipitation and global meteorological situation influenced by sunspot cycles (Hunyadi *et al.*, 1988).

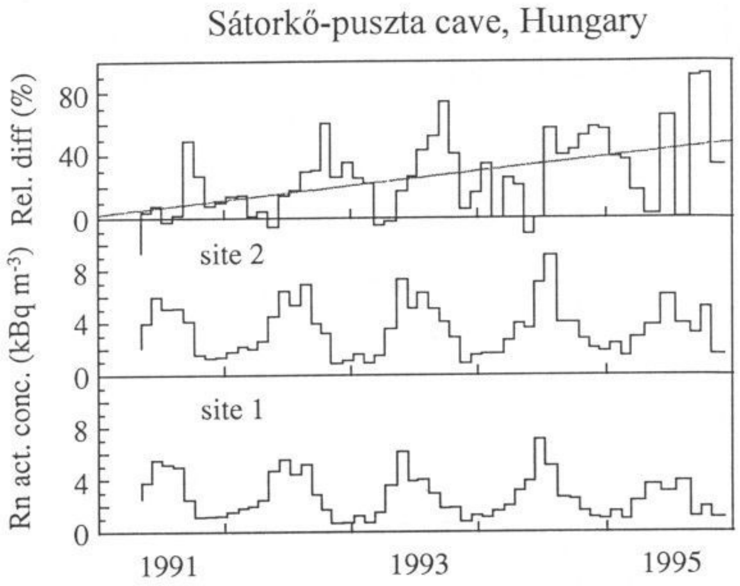

Figure 4.1.4: The radon activity concentrations at two measuring sites and the relative difference between them representing the Sátorkő-puszta cave, Pilis Mountains, N Hungary.

4.1.6 Radon as a Natural Radioactive Tracer of Subsurface Transport Processes

Subsuface natural fluids in the majority of cases carry small amounts of environmental isotopes. The behaviour of these elements, and the variation of their concentration in time and space, is the result of physical, chemical and biological interactions. These elements whose physical properties, concentrations, etc., provide information on flow and kinetics of the carrying substances are called natural tracers.

The application of radon as a natural tracer is not yet common and widespread. Among the natural tracers it would be considered on the one hand as ideal, since it is easily detectable even in small quantities, and does not modify the characteristics of the environment. On the other hand, unfortunately its sources appear everywhere and are spread over in a manner unknown *a priori.* Therefore the interpretation of the concentration data is not straightforward: it needs interdisciplinary expertise of hydrogeologists, geologists, physicists, radiogeochemists, etc. Joint efforts have given results in different fields. The observations of subsurface fluid motions traced by natural radon were followed by new ideas about the basic transport phenomena and, later, by new interdisciplinary applications: as, for example, mapping of active faults; investigations of volcanic and seismic activities; earthquake prediction; hydrogeological research, etc. (Fleischer, 1988).

In the speleology, similarly to the previously mentioned fields, these types of measurements have already found their applications, and they make important contributions to the better understanding of the natural regimes of caves. Cunningham and LaRock (1991) delineated six microclimate zones in Lechuguilla cave, Carlsbad caverns, National Park, New Mexico, using radon grab sampling in conjunction with observed air flow data. Atkinson *et al.* (1983) from a single set of etched track measurements in the Castleguard cave, Columbia Icefields, Alberta, Canada, identified the effect of tributary air currents from larger fissures.

The most common and most apparent phenomenon which was discovered in the majority of the investigated caves throughout the world was the annual change of radon activity concentration. Wilkening and Watkins (1976) identified temperature gradients favourable to vertical convective transport through relatively large openings. They identified as well transport of radon by air movement through cracks and fissures due to pressure gradients (Wilkening, 1980). As karstic caves are situated generally in highly fracturized rocks, such a configuration is favourable for the emergence of air circulation through this fracture system. The strength of such air motions is taken to be proportional, to a first approximation, to dT/f, where dT is the temperature difference between the cave and outside and f is a friction factor characterizing the flow resistance (Atkinson *et al.*, 1983; Quinn, 1988; Wilkening and Watkins, 1976). A typical radon time-series indicating the temperature control is shown in Fig. 4.1.2 (Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary). On approaching deeper parts of the karst, the radon levels are mostly stable, as the strength of air motions decreases due to their 1/f dependence; and owing to saturation effects the amplitude of the changes also decreases (Hakl *et al.*, 1992). Radon levels in deep, complex caves cannot be simply related to outside atmospheric parameters (Cunningham and LaRock, 1991).

The actual value of the radon concentration in the cave is influenced by subsurface fluid motions due to the periodically or randomly changing gradients in the environmental parameters (temperature, pressure, humidity, stresses, ...), and by the radon concentration saturated in the pore spaces of the surrounding rocks. Using long-term temporal and large-scale spatial variation measurements from the analysis of temporal radon variations, the complex interplay of the two traced substances, water and air, can be identified. Such measurements were performed in the Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary (Lénárt *et al.*, 1988). Typical patterns of temporal changes are characterised by summer maxima and winter minima. Similar variations were found at almost all measuring sites, with the exception of that part where more or less continuous water inlets were present. According to the temporal variation, Lénárt *et al.* (1988) differentiated three parts in the cave. In the first part (entrance part) the mean radon activity concentration in air increased from 1 kBq m^{-3} to 2.2 kBq m^{-3}. In the second part it fluctuated around 2.2 kBq m^{-3}. We note that in these two parts there was a slight asymmetry in the way the radon level changes between summer and winter. In the springtime, radon levels in the cave increase gradually, whereas in the fall they decline rapidly. (On increasing the time resolution of the measurements, the same effect can be made much more pronounced, as was found by Lively and Krafthefer (1995) using active techniques.) The highest air radon values, around 4 kBq m^{-3}, were found in the third part, which was characterised by the presence of several streams and a Lake fed by them. In the third part the dominant role of the periodic stream is evident from the long-term measurement (see Fig. 4.1.5); while in the first two parts the change of the radon activity concentration showed close connection with the temperature difference between the cave and outside.

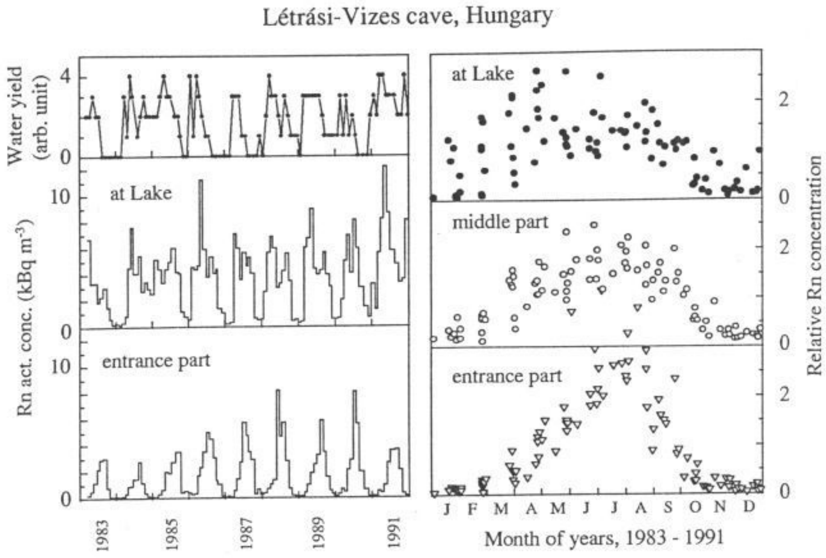

Figure 4.1.5: The yield of the stream feeding the Lake and the temporal change of radon activity concentrations in air at the Lake and at the entrance part of the Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary (left). The form of the upper curves, in spite of low temporal resolution, shows a connection between the two quantities. The phenomenon is explained by the fact that water is the dominating radon source in cases of floods. The difference in springtime variations of normalized monthly radon distributions (right) reflects the same phenomenon, simultaneously showing radon regimes of the three distinct microclimatic zones.

Radon levels in some cases show significant correlation with barometric pressure, but as the characteristic time of pressure changes is mostly less than the exposure time of track detectors, this type of phenomenon can be revealed only by active methods.

4.1.6.1 Modelling of observed features

The underground radon transport can be described by the following transport equation

$$\frac{\partial C}{\partial t} = D_{eff}\Delta C - \nabla(\vec{v}C) - \lambda C + \phi \qquad (4.1.1)$$

where C [m^{-3}] is the radon concentration in pore space, D_{eff} [$\mathrm{m}^{-2}\ \mathrm{s}^{-1}$] is the effective diffusion coefficient of radon, $\vec{v}$ [m] is the velocity of the carrying substance, λ [s^{-1}] is the decay constant of radon and ϕ [$\mathrm{m}^{-3}\ \mathrm{s}^{-1}$] is the source term. In the equation, the first term describes diffusion, second term convection, third term decay and fourth term radon sources. For the solution of transport equation, first of all it is necessary to know the velocity field (e.g. Navier-Stokes equation, which by itself is a sufficiently complicated problem); then taking into account the source term, and the initial and boundary conditions C can be determined. The emerging phenomena are determined by the form, shape and structure of the underground void space. The above equations can be generally solved only numerically.

The realisation of radon transport processes sharply depends on the configuration of the interconnected underground cavities. In the case of blind-end systems, atmospheric pressure changes are the main control parameter (Ahlstrand, 1980; Hakl *et al.*, 1995; Wigley, 1967), which are superimposed by convective air exchange due to temperature differences in cave systems with vertical extension. In case of relatively large entrances the convective air exchange due to temperature differences can mostly account for the radon transport process taking place (Wilkening, 1979). In the case of two or more entrance systems, where the other 'entrances' can be complexes of smaller fissures and fractures, chimney effect winds may dominantly govern the radon transport; or in some cases atmospheric winds may do so.

The interpretation of the data can be affected by the presence of these unknown (unassumed) 'entrances'. Yarborough (1980), in a study of nine caves, identified two general types of physical cave configurations that affect airflow patterns and radon concentrations. Type 1 caves have most passages at or above the entrance elevation; Type 2 caves have most passages below the elevation. When the outside temperature exceeded the cave temperature, he found that Type 1 caves exhaled, while stagnation occurred in Type 2 caves; when the outside temperature fell below the cave temperature, both types inhaled. As Type 1 cave is a horizontal mirror of Type 2 cave, the seasonal phenomena should be opposites of each other, so stagnation should occur in Type 1 cave in winter time. Winter time stagnation was identyfied in Pisznyice cave, Gerecse Mountains, Hungary, resulting in high radon concentrations in the cave during the cold season (Hakl, 1994).

The above examples show the problems of interpretation and modelling. In a few special cases, however, the emerging processes are analytically surveyable. These are the cases of idealised two-entrance horizontal flow-through, and one narrow entrance vertical caves. In the case of the horizontal model cave, the second entrance may represent the set of vertical fractures which connect the main passage to the surface through the overburden.

First, let us consider a schematic horizontal cave (see Fig. 4.1.6, left). In this case, owing to the temperature dependence of air density the pressure exerted at the lower end of the cave by the outside air will differ from that inside. This pressure difference in this case is (Atkinson *et al.*, 1983)

$$\Delta p \approx -gh\rho_{in}\frac{\Delta T}{T_{out}} \tag{4.1.2}$$

where ρ_{in} is the density inside air, g is the gravitational acceleration, h is the height difference between the two entrances, ΔT is the temperature difference between the cave and the outside, and T_{out} is the outside temperature. According to this relation, the direction of air flow through the entrance depends on the season; in warm season air flows out of, and in cold season air flows into, the cave.

In the case of narrow-entrance vertical caves, the most effective processes in inducing air motions are the atmospheric pressure changes (Fig. 4.1.6, right). Falling ambient atmospheric pressure drains air from the cave; increasing atmospheric pressure presses air into the cave through the entrance. The volume of air passing the cave entrance

$$\Delta V \approx -\frac{V_{cave}}{p_{out}}\Delta p \tag{4.1.3}$$

where V_{cave} is the cave volume, p_{out} and Δp are the ambient atmospheric pressure and the change in the ambient atmospheric pressure, respectively.

We note that air flows induced by atmospheric pressure changes can be rather quick. At the entrances of giant caves their speed can reach several tens of km h^{-1}.

The above considerations may suggest that advection is the main physical process, which governs the radon transport. This implication depends, however, on the size and density of openings and fractures, as they play an important role in the forming of radon concentrations as well as in the dynamics of transport processes inside them (Hakl, 1993; Scherry and Siegel, 1986). The effect of geometrical parameters can be well investigated in a three-dimensional model of a cylindrical void embedded in the rock matrix. By solving the appropriate transport equation, the transport length (z) may be derived as the parameter which characterises the radon transport inside the cylinder

$$z = \frac{v}{\lambda}\frac{1}{1+2(z_d/r_0)R(r_0)} \tag{4.1.4}$$

where v is the air flow velocity, λ is the decay constant of radon, z_d is the effective radon diffusion length in rock matrix, r_0 is the radius of the cylinder, and R is the negative of the ratio of the derivative of the zeroth order modified Bessel function to the zeroth order Bessel function ($i.e. R = -K_0'/K_0$) evaluated at r_0/z_d.

According to this formula two extreme cases can be distinguished: (a) In the region where $z << z_d$, diffusion dominates as the radon concentration of the moving air is adapted to its bounding environment at very short distances; and (b) If $z >> z_d$, advection dominates: radon is transported much more quickly than is characteristic for diffusion.

Physically, the above two conditions mean that the convective processes taking place along the underground openings, and the diffusive processes perpendicular to the walls of channels, compete with each other. The critical speed v_{crit}, of a flowing substance in a channel, therefore, can be defined in such a way, that the characteristic transport lengths in porous matrix and in the channel equals with each other, i.e.

$$\sqrt{\frac{\lambda}{D_{eff}}} = \frac{\lambda}{v_{crit}}\left(1+2\frac{z_d}{r_0}R(r_0)\right) \tag{4.1.5}$$

Substituting typical values for diffusion length for limestone (0.2-0.4 m) yields $v_{crit} = 0.2$-20 mm h^{-1}, depending on the size of the channel (0.002-2 m).

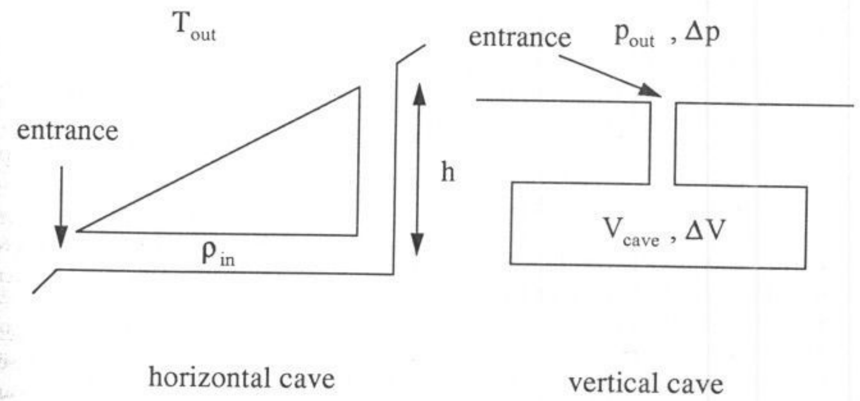

Figure 4.1.6: Schematic horizontal and vertical caves; h is the elevation difference between the upper and lower entrances (horizontal cave), ρ_{in} is the air density of inside air, T_{out} is the outside temperature, p_{out} is the ambient outside pressure, V_{cave} is the cave volume and ΔV is the volume of the air passing the vertical cave entrance in case of Δp change in the ambient outside pressure (not to scale).

A more realistic model of fracturized karstic overburden above a horizontal cave is the model of parallel voids connecting the main passage to the surface (Géczy *et al.*, 1989; Scheidegger and Liao, 1972). This model is a self-evident synthesis of the idealised two entrance horizontal cave model and cylindrical void embedded in the rock matrix model. In the warm season, according to the Δp pressure difference, air will flow into the cave from the direction of the fracture system (and will flow out of the cave through the entrance at lower altitude). If the velocity of the flow is 2-3 times higher than the critical value, the flow will be able directly to wash out radon from the fractures. If the velocity is less than the critical velocity, then the diffusion profiles in the rock matrix are shifted in the first step, and therefore increased radon flux can reach the cave passage only with delay. The level and temporal variation of radon concentration in the cave gallery therefore depend on the density and size of the fractures. In the cold season, the direction of the flow is reversed. From the direction of the lower entrance fresh outside air flows into the cave forming considerably lower radon concentration levels.

By the application of etched track technique alone, the above-mentioned two competitive processes can hardly be discriminated between, owing to the limited time-resolution of etched track technique. On applying the combination of active electronic and passive etched track techniques, however, most of the hidden phenomena can be discovered (see subsubsection 4.1.8.1).

4.1.7 Radon Risk Inside and Above the Caves

It is known that high radon exposure results in excess lung cancer mortality rate among miners. A majority of experts believe that elevated radon levels in homes induce excess lung cancer mortality rate among the general public, too. High radon levels that occur in caves may raise the question whether any people are at risk or not. Most concerned groups are the cavers and tour guides. Moreover, some cold karstic caves are used for therapeutic treatment of patients suffering from chronic respiratory diseases. These patients, and the staff members of the therapy teams, are also concerned about being subjected to higher risks. Hungarian data were summarised by Csige *et al.* (1996) (see Table 4.1.1). Currently, in Hungary radon is continuously monitored in all the five caves where therapeutic treatment of bronchial asthma and chronic bronchitis is going on. Some caves open to tourists are also monitored. In the case of the best known of these (Baradla cave, Aggtelek Karst, Aggtelek National Park, Hungary), the radon doses of tour guides were estimated for the years 1990-1994.

According to Navrátil *et al.* (1994) the range of patients' doses from nine different therapeutic caves in Europe is 0.07-1.32 mSv, which well fits with the above table. Based on internal working time reports, annual effective doses for therapy staff members are about 0.4 mSv in Abaliget cave, 0.12 mSv in Szent István cave and 6 mSv in Béke cave.

Annual personnel exposures also cover a wide range. Tour guides in Abaliget cave receive about 12 mSv annually, Cigna and Clemente (1981) cite Yarborough, who has found 0.005-19.9 mSv per year for seven different US caves; whereas Nikodemová (1995) reports effective dose rates 0.17-4.05 mSv per month for personnel in Slovakian caves.

Table 4.1.1: Summary of radon doses received on the course of speleotherapy in Hungary (Csige *et al.*, 1996).

Cave name	Period	Sample size	Annual effective dose (mSv) Mean, (Range)	Annual effective dose (mSv) Geometric mean, (Geometric STD)
Abaliget[1]	1994	127 patients	0.54, (0.03-1.26)	0.40, (0.77)
Béke[2]	1994 summer	56 patients	1.91, (1.86-1.97)	1.91, (0.02)
Szemlőhegy[*,3]	1990-1992	229 patients	0.85, (0.10-5.00)	0.62, (0.76)
Szent István[2]	1994	360 patients	0.06, (0.01-0.17)	0.04, (0.88)
Tapolca[4]	1994	481 patients	0.87, (0.04-2.19)	0.45, (1.32)
Baradla[2]	1990-1994	12 tour guides	2.66, (0.12-5.55)	2.13, (0.80)

[1] - Mecsek Mountains, S Hungary;

[2] - Aggtelek Karst, Aggtelek National Park, NE Hungary;

[3] - Buda Mountains (at Budapest);

[4] - Balaton Highland;

[*] - In the case of Szemlőhegy cave cumulative effective doses are given over 1990-1992, as not all the patients attended the therapeutic courses each year.

Most active cavers may be exposed to even higher doses; so personal radon dosimetry is highly recommended for those people. One of the authors (J. Hakl) has measured his own annual effective dose due to radon inhalation while working in different caves, and found that it was higher than 30 mSv in 1992. Even higher personal doses can be received in caves with unusually high radon concentrations. Hyland and Gunn (1994) estimated that it took 33 hours to reach 15 mSv limit in some caves of North Pennines. Two important radiological consequences follow from the periodical behaviour of subsurface air circulation in karst. First is the seasonally varying underground radon level which results in a large difference between summer and winter values, and hence also in the radon exposures received by cave visiting people.

The other consequence of the seasonally directed transport phenomenon is that variation in radon exhalation can also be expected on karstic terrains seasonally. A very convincing experimental result of the phenomenon is represented in Fig. 4.1.7, where radon time-series measured inside the Hajnóczy cave, Hungary (with minima in the cold winter season), and in a slit above the cave (with winter maxima) are shown. Similar winter maxima were found on several karstic terrains of Hungary (Hakl *et al.*, 1992). Elevated radon levels already observed in houses in summertime (Wilson *et al.*, 1991) and wintertime (Gammage *et al.*, 1992) are attributed to this phenomenon, showing that the season of maxima depends on the position of the house with respect to the underground air circulatory system. The inverse correlation of radon levels inside a cave and in a house above the cave was found directly by Lively and Krafhefer (1995).

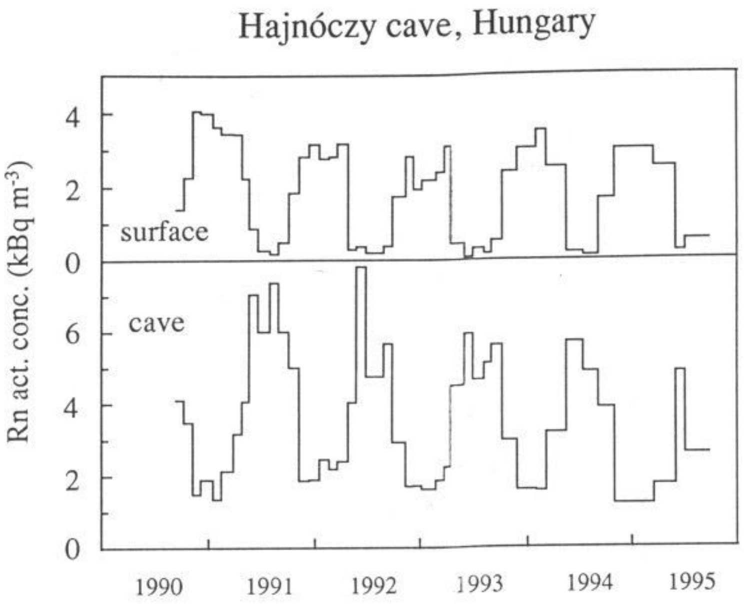

Figure 4.1.7: Push-pull type change of seasonal radon activity concentrations observed in the Great Hall of the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary, and in a surface slit above the cave.

4.1.8 Future Developments

4.1.8.1 Combination of etched track and real-time detection techniques

Combining etched track and real-time detection techniques, spatial and detailed temporal variation of radon concentration can be obtained. The information content of data can be increased substantially, but the choice of proper measuring places is also desirable. By choosing proper and right number of test sites, the functioning of the subsurface system can be understood in greater detail. Such type of combination of continuous and integrating radon measurements was performed in the Vass Imre cave situated in the Aggtelek Karst, Aggtelek National Park, NE Hungary. This cave is very similar to the model cave surmised by the air circulation model. It has one entrance, and is situated practically horizontally with no visible vertical connection to the surface. In the middle of the cave there is a syphon, which was closed by water several times during the observation period (1987-1993). When the syphon is open, there is continuous air flow either in or out from the cave through the entrance (winter and summer, respectively). When the syphon is closed, there is no measurable air flow through the entrance.

Table 4.1.2: Mean radon activity concentrations of air (kBq m^{-3}) in the Vass Imre cave, Aggtelek Karst, Aggtelek National Park, NE Hungary (1987-93).

	Before syphon		After syphon		Annual mean
	Winter	Summer	Winter	Summer	
Syphon opened (air flow exists)	1.2	8.5	1.2	8.5	4.8
Syphon closed ('no' air flow)	2.5	4.0	3.5	3.1	3.3

The radon concentration is being measured with etched track detectors since 1987 at 12 sites located at equal intervals along the cave passage, and in 1991 three more continuous real-time radon monitors were installed at characteristic points of the cave.

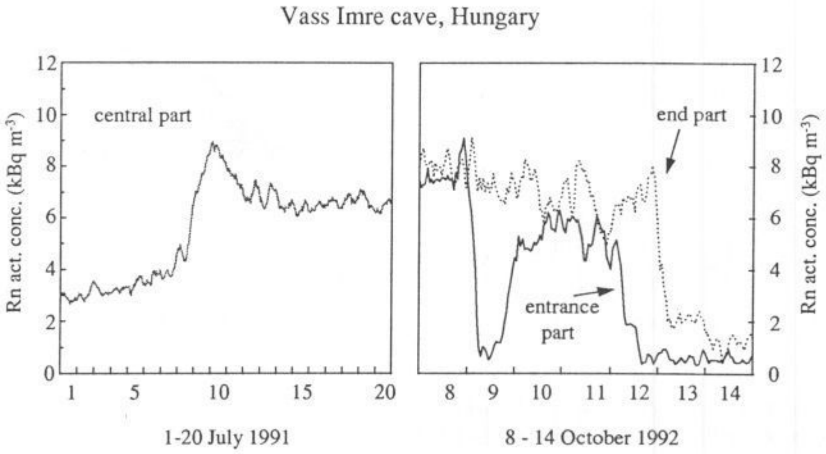

Figure 4.1.8: The real time records of radon time-series from Vass Imre cave, Aggtelek Karst, Aggtelek National Park, NE Hungary, at the times of syphon opening (left) and arrival of early cold fronts during fall (right).

Etched track detectors changed monthly showed elevated levels in the summer and lower levels in the winter: the temporal change is of a smooth sinusoidal type. The horizontal radon distributions along the main cave passage were uniform. The characteristic radon values found are summarised in Table 4.1.2.

The penetrability of the syphon affects markedly the mass of infiltrated air through the cave system by controlling the strength of the emerging air flows. When the syphon is closed, the restriction of the air flows has a different effect on the forming of radon levels, which depends on the season. In summertime the restricted air flow decreases, but in wintertime it increases, the mean radon levels in the cave, resulting in an overall drop of 30% in the annual mean radon concentration (see Table 4.1.2). This asymmetric effect can be explained on the basis of the air circulation model (Géczy *et al.*, 1988) and is in agreement with the results of numerical calculations (Holford *et al.*, 1993) showing the increase of mean radon levels due to periodically changing flow conditions.

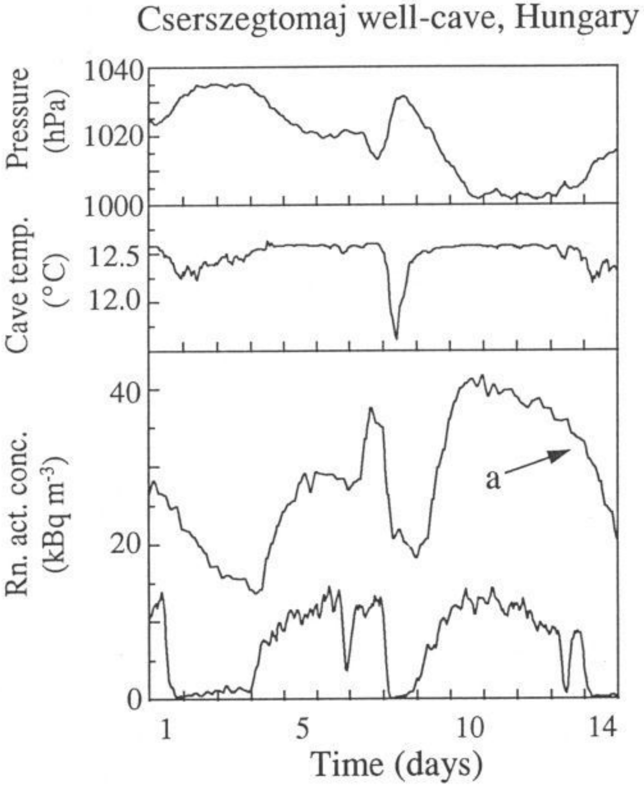

Figure 4.1.9: Temporal variation of temperature, pressure and radon activity concentration at the entrance part of the Cserszegtomaj well cave, Keszthely Mountains, SW Hungary. On the figure we also displayed the change of radon level measured deeper inside the cave (a).

On analysing temporal changes in radon time-series, we found advection as the dominant transport process. This is also shown on the transient part of the radon record from the Vass Imre cave, Aggtelek National Park, NE Hungary, which corresponds to a summertime syphon opening (see Fig. 4.1.8 left). The shape of the curve, on comparing it to the results of numerical calculations on radon exhalation from porous media (Janssens *et al.*, 1984), corresponds to the case when a sudden pressure drop results in instantaneous air-flow development. The pressure drop effect was also observed experimentally during a planned syphon closing and opening (Kérdő, 1994). On the other hand, the absence of strong daily changes in the radon record, which would correspond to the observed daily air flow fluctuations (Holl, 1993), shows the strong smoothing effect of diffusion, which can be due to relatively undeveloped fracture system of this cave. (We note that in caves located in more karstified environment, more or less pronounced daily fluctuations in radon records can be observed.)

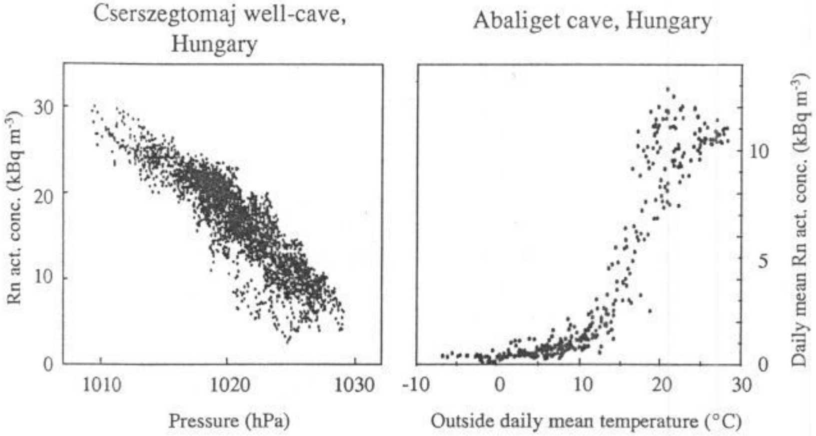

Figure 4.1.10: Radon activity concentration vs atmospheric pressure observed at one of the five continuous observation stations in the Cserszegtomaj well-cave, Keszthely Mountains, SW Hungary (left). The 'data cloud' corresponds to the 2 month section of the continuous data record. The inverse pressure control on radon levels is well seen in the figure. On the right-hand part of the figure we have displayed radon levels vs outside temperature observed in the Abaliget cave, Mecsek Mountains, S Hungary. The S-type dependence of radon concentration on the outside temperature is typical of caves dominated by chimney-effect winds.

Similar conclusion can be drawn by analysing passive etched track radon measurements. Because the radon concentration after the closed syphon does not depend on the season, the radon diffusion length in the rock matrix can be estimated, taking into account the geometry of the cave passage. Knowing the diffusion length, the air-flow velocity through the fracture system can be further appraised, using the summer data. The resulting speed is in the range (0.5-1)$\cdot 10^{-4}$ cm s^{-1}. This speed is less then the critical speed, which points to the diffusion control, as was deduced from analysing the real-time data.

In contrast to the above phenomena, the cave radon concentration 'shut down' is very quick in autumn, and is controlled purely by advection. An early cold front of short duration caused a fall in radon records at the first and second measuring sites, but it did not affect the end of the cave. High radon levels were recovered for two days by reversing the air-flow. Finally, permanent low radon values were formed in the cave air after the arrival of another, stronger, cold front. The time-difference between the radon falls corresponded to an air transport velocity of about 50 m h^{-1} along the main cave passage (see Fig. 4.1.8, right).

In some case, in caves with small entrances the changes in outside temperature have only a small effect on radon concentration inside the cave, and only on an annual scale. In contrast, the atmospheric pressure has strong influence on radon level: pressure changes play an important role in controlling radon transport. The radon time-series, observed in the Cserszegtomaj well-cave, Keszthely Mountains, SW Hungary, show this effect markedly (Fig. 4.1.9). Radon transport is predominantly controlled by advection, as is shown by the immediate response of radon concentrations to pressure changes. Decreasing pressure increases the radon level and conversely. In response to the changes in atmospheric pressure, radon 'pulses' can be observed between monitoring locations. The figure also shows the difference in phases of the site-specific radon responses. The hysteresis found in the pressure - radon correlation curve indicates nonlinearity of processes (see also Fig. 4.1.10, left). It corresponds to a time shift in the response of air-flow velocity to the pressure changes (Wigley *et al.*, 1967). From the data it is also possible to estimate the cave volume, using simultaneous pressure measurements (and utilising the known volume of the vertical entrance well). We found it to be 8-10 times larger than was estimated form the calculated volumes of passages.

In the Fig. 4.1.10 (right) we have, for the sake of comparison, also displayed the radon response of a chimney-effect controlled cave system.

4.1.8.2 Radon monitoring network

From the previous subsections it is evident that, from radon records, more information can be deduced unambiguously, taking account of other environmental parameters. This type of multiparameter approach is an emerging indispensable task in environmental transport studies.

As the role of the geological environment and the necessity of multiparameter studies has recently been recognised, there is an increasing need for instruments suitable for long-term field measurements. Owing to the complexity of the radon transport problems, these above-mentioned studies require numerous, continuously operating stations. From the scientific and economic point of view, an independent working ability is more and more frequently required (the usual on-line measurement and data transmission in real time are prohibited by cost and insufficient infrastructure).

These efforts initiated, in Hungary, the development of the prototype of the Dataqua field multiparameter radon monitor in the beginning of the 1990's, which opened new dimensions in radon research. In many places in Hungary, such complex radon monitoring stations have now been installed. At the test sites, radon concentrations together with other environmental parameters are continuously recorded with a sampling frequency of one measurement per hour. The main aims in operating these monitoring stations are:

- to indicate whether or not there are extremely high radon concentrations in places where people live or work (dwellings, cellars, caves, coal mines, uranium mines),
- to obtain radon output activity of some objects of interest (uranium mine, mill and tailings, coal mine and power plant, coal ash tailings, etc.) into the environment,
- to study the relationship between the variation of radon concentration and other environmental parameters (air temperature, barometric pressure, water level variation, etc.),
- to reveal the connection - if any - between radon exhalation and geodynamical events (earthquakes, fault movements, rock explosions in the mine, etc.).

The first field results proved the usefulness of the radon monitoring system. The advantage of this system, in comparison with the traditional solid state nuclear track detector technique alone, is obvious: the time resolution of observations has revealed previously unknown details of temporal variations, and by interpreting them one can define new approaches of physical models describing the objects of interest. Some of the results have been highlighted in foregoing sections. We note, however, that enormous efforts (including software and hardware) are required to analyse and interpret the great amount of time-series recorded (some 10,000 data per year per monitor).

4.1.9 Concluding Remarks

Research activity has been developed in the field of geologically related radon transport studies. As radon is an easily measurable radioactive element, it is a very convenient natural tracer for the transport processes taking place at the interfaces of the atmosphere, hydrosphere and lithosphere. It brings information from the depths of the earth, which may be helpful in the prediction of earthquakes, lava eruptions and other geodynamical events as mine outburst. Because of the improved time resolution, the proportion of active automatic radon measuring methods is continuously increasing in this field.

By interpreting the observed long-term radon time series, new results will be obtained from intercomparison of the dynamical behaviour of radon concentrations at different geographical and geological sites. In this way, general understanding of environmental (radon) transport processes will be improved.

The measurements in karstic caves will rely on examining the natural ventilation regimes. Because the interrelations between surface and cave environments are affected by flowing air masses, the exploration of transport processes is a precondition to gathering knowledge about the development and changes in the cave microclimate and about the propagation mechanism of external effects. In this way, these results should also find application in the protection of the karst environment.

The radon values should be compared with environmental parameters, e.g. microclimate, temperature, humidity, ventilation rates etc. The spin off would be to understand emanation control in these environments, and as a result this would help in controlling the radiation hazard.

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RADON TRANSPORT PHENOMENA STUDIED IN KARST CAVES - INTERNATIONAL EXPERIENCES ON RADON LEVELS AND EXPOSURES

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ABSTRACT

The results of radon measurements in caves obtained by using of nuclear track detectors are summarized. Mean radon concentrations are ranging worldwide from 0.1 to 20 kBqm^{-3} with 2.8 kBqm^{-3} arithmetic average. From long-term extended radon measurements in caves not only a detailed dosimetric picture can be drawn, but using radon gas as a radioactive tracer, the subsurface and near-to-surface transport processes can be studied, too. It will be shown that long-term radon monitoring by nuclear track detectors, in conjunctions with active detectors which enables detection of fast dynamic changes, offers very important information for naturally-occurring transport processes.

KEYWORDS

Radon; cave; natural tracer; karst; etched track;

INTRODUCTION

Limestone contains in average about 1.3 - 2.5 ppm ^{238}U (16 - 31 Bq·kg^{-1}), e.g. its daughter product ^{222}Rn can be found naturally in all caves. These minute quantities of parent substance result in relatively high values of radon in caves. Caves occur everywhere on the earth, although mainly in karst areas - as they are formed mostly in limestone environments. Karst strata have a characteristic feature: due to the infiltrating waters a special chemical dissolution process is taking place in the rock matrix. The phenomenon results in forming of an interconnected set of larger openings and fractures; a cave is a part of this system. Due to the formed special morphology the cave is able to communicate through the overburden under the influences of changing atmospheric pressure or temperature. These morphology, meteorology (and surface topology) linked flows play the most significant role in governing the underground radon transport in these areas.

In the last two decades the etched track techniques have found their place also in radon measurement projects aimed to study the cave environment. These measurements have ranged from sporadic observations up to regular long-term studies, and they were motivated by dosimetric and transport study approaches as well. In Hungary the first *in situ* field radon measurements in caves performed by

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etched track techniques started as early as 1977 and were initiated by Dr G. Somogyi, one of the pioneers of the etched track field. Since then, the number of studied caves has increased substantially and we have reached 10000 observation data in 31 Hungarian caves. After recognizing the link between the secondary porosity, fracturization and formed radon levels the ever-pressing interest to understand underground radon transport in karst environment has been increased. During the last several years active electronic devices, automatic multiparameter radon monitors have been included into cave studies. Including a short literature review the following results are excerpts based on this data set.

MATERIALS AND METHODS

For the purpose of cave radon measurement, different types of opened and closed diffusion chambers, equipped with alpha sensitive polymer track detectors, were used. Somogyi *et al.* (1983) described different types of multi-detector devices suitable for measurement in caves. The most significant improvement in the measuring technique was the thermally stabilized double-wall diffusion chamber (filled with water), which avoided the frequent problem of wetting of detector surfaces. Nowadays several commercially available radon monitors developed for indoor radon measurements are used in cave environment, which, owing to the high relative humidity, may be not always optimal. The typical exposure time ranges from one week to several months.

RESULTS AND DISCUSSION

Review of Published Data

Continuous radon measurements covering at least one year long period were performed by the etched track technique in Hungary, Italy, Slovakia and Luxembourg. Seasonally one week long observations were performed in England, and measurements not covering a full year are reported from Mexico and USA.

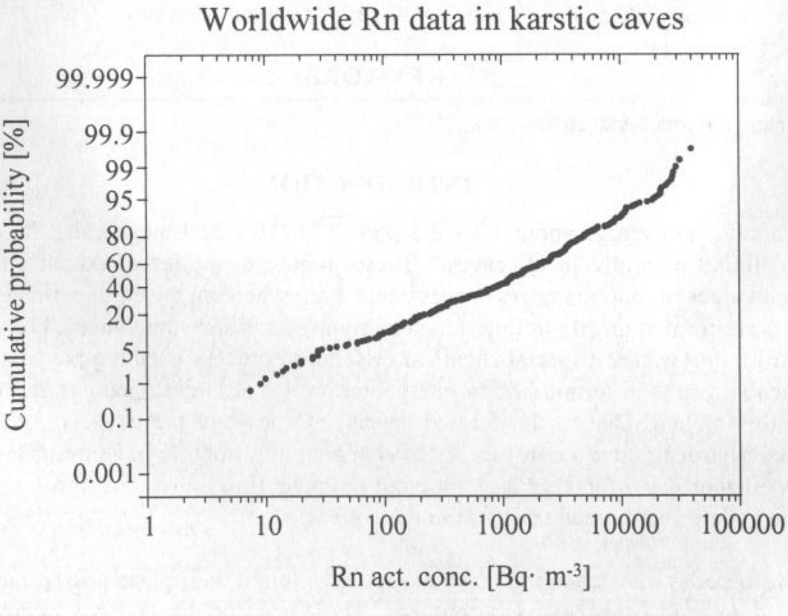

Figure 1. The distribution of radon activity concentrations reported from different caves worldwide. In the graph we compiled radon data available from 220 different caves.

Taking into account these results the annual average radon concentrations in karst caves range from 0.1 to 20 kBqm^{-3} with a 2.8 kBqm^{-3} arithmetic average. The lower end of the scale is associated with big chamber volumes or high ventilation rates; the upper end is characterized by closed, badly ventilated places and uranium-rich sediments. We note however, that based on long-term continuous radon observation the relative variations of radon between monitor locations indicate that the cave atmosphere is not a uniform radon environment over any given time period; so it is very hard to represent one cave with one radon value. The distribution represented on Fig. 1. provides compiled results of cave radon literature. The distribution of the data is close to lognormal.

Radon as a Natural Radioactive Tracer of Subsurface Transport Processes

The application of radon as a natural tracer is not yet common and widespread. Among the natural tracers it would be considered on the one hand as ideal since it is easily detectable even in small quantities, which do not modify the characteristic of the environment. On the other hand, unfortunately its sources appear everywhere and are spread over in a manner unknown *a priori*. The realization of radon transport processes in addition sharply depends on the configuration of the interconnected underground cavities. In the case of blind end systems, atmospheric pressure changes are the main control parameter, which are superimposed by convective air exchange due to temperature differences in cave systems with vertical extension. In the case of relatively large entrances, the convective air exchange due to temperature differences can mostly account for the radon transport process taking place. In the case of two or more entrance systems, where the other 'entrances' can be complexes of smaller fissures and fractures, chimney effect winds may dominantly govern the radon transport, or in some cases atmospheric winds may do so. It is obvious, that the interpretation and modeling of the concentration data is not straightforward: it needs interdisciplinary expertise of hydrogeologists, geologists, physicist, radiogeochemists, etc.

In the speleleology these types of applications give important contributions to the better understanding of the natural regimes of caves. Cunningham and LaRock (1991) delineated six microclimatic zones in Lechuguilla cave, Carlsbad caverns, National Park, New Mexico using radon grab sampling in conjunction with observed airflow data. Atkinson *et al.* (1983) from a single set of etched track measurements in the Castleguard cave, Columbia icefields, Alberta, Canada, identified the effect of tributary air currents from larger fissures.

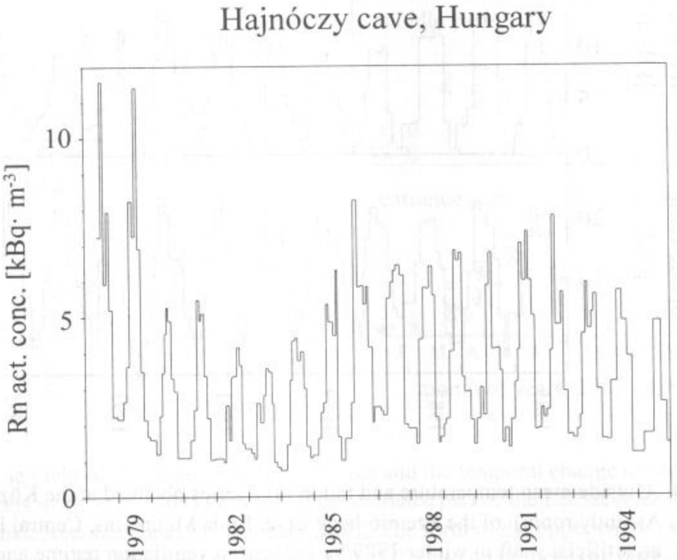

Figure 2: Radon time series observed by using etched track technique in the Nagy-terem (Great Hall) of the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary. Each observed value is the average of three simultaneous readings with one month integration time.

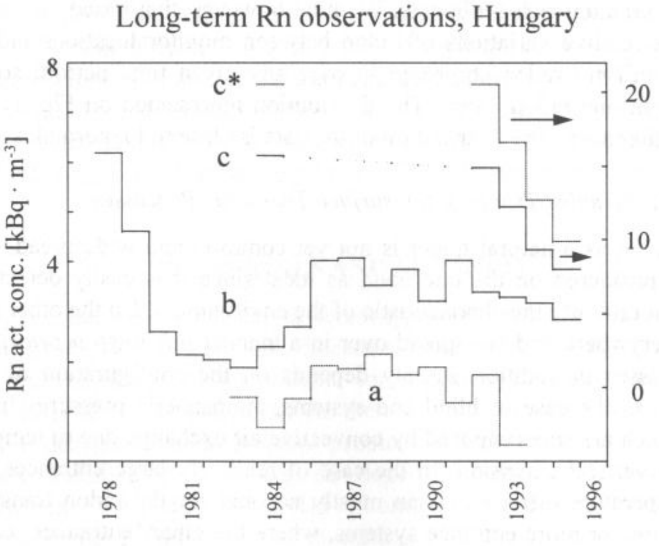

Figure 3: Long-term change observed in three Hungarian caves of different types. Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary, is dominated by chimney effect winds; Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary, is characterized by temperature difference driven convective air motions; and Cserszegtomaj well-cave, Keszthely Mountains, SW Hungary, is influenced mainly by atmospheric pressure changes. The curves represent the middle part of the Létrási-Vizes cave (a); the Nagy-terem (Great Hall) in the Hajnóczy cave (b); and cave average (c) and a remote part (c*) in the Cserszegtomaj well-cave.

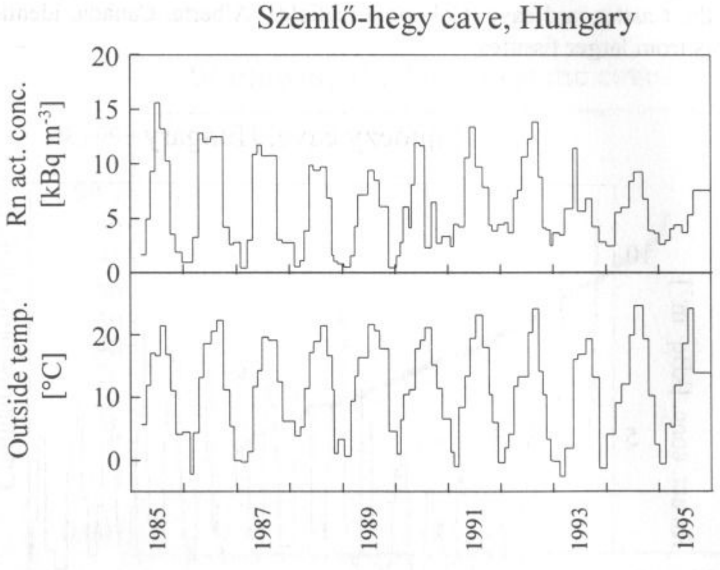

Figure 4: Outside mean temperature and radon time series obtained at the Közgyűlés-terem (General Assemly room) of the Szemlő-hegy cave, Buda Mountains, Central Hungary. The sealing of an artificial shaft in winter 1989/90 influenced ventilation regime and consequently radon level in the cave.

So far the longest time series were obtained in the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary (Fig. 2), which shows very regular seasonal variation. The same effect was found in a few other Hungarian caves, but it is also reported by Lively and Krafthefer (1995). The observed fluctuations of smaller or larger amplitude are a general phenomenon, where the frequency and amplitude distribution of the data is a common characteristic for the cave and its environment.

It is also clearly seen on the Fig. 2, that the seasonal variation is superposed on a long-term change of the mean activity concentration. This long-term change can be found in different types of caves, as it is shown on Fig. 3. They display the effect of slowly changing environmental parameters on radon transport processes. Such external parameter may be the change in radon emanation power due to the slow change of water content of the cave surrounding, which may reflect the variation in annual precipitation and global meteorological situation influenced by sunspot cycling (Hunyadi *et al.*, 1988).

The observed annual change of radon activity reflects two basic processes. Wilkening and Watkins (1976) identified temperature gradients favorable to *vertical convective transport* through relatively large openings. They identified as well transport of radon by *air movement through cracks and fissures* due to pressure gradients (Wilkening, 1980). As karst caves are situated generally in highly fracturized rocks, such a configuration is favourable for the emergence of air circulation through this fracture system. The strength of such air motions is taken to be proportional, to a first approximation, to dT/f, where dT is the temperature difference between the cave and outside and f is a friction factor characterizing the flow resistance (Wilkening and Watkins, 1976; Atkinson *et al.*, 1983). A typical radon time series indicating this type of temperature control is shown on the Fig. 4 (Szemlő-hegy cave, Buda Mountains, Central Hungary). On approaching deeper parts of the karst, the radon levels are mostly stable, as the strength of air motion decreases due to their 1/f dependence; and owing to saturation effects the amplitude of changes also decreases (Hakl *et al.*, 1992). Radon levels in deep, complex caves cannot be simply related to outside atmospheric parameters (Cunningham and LaRock, 1991).

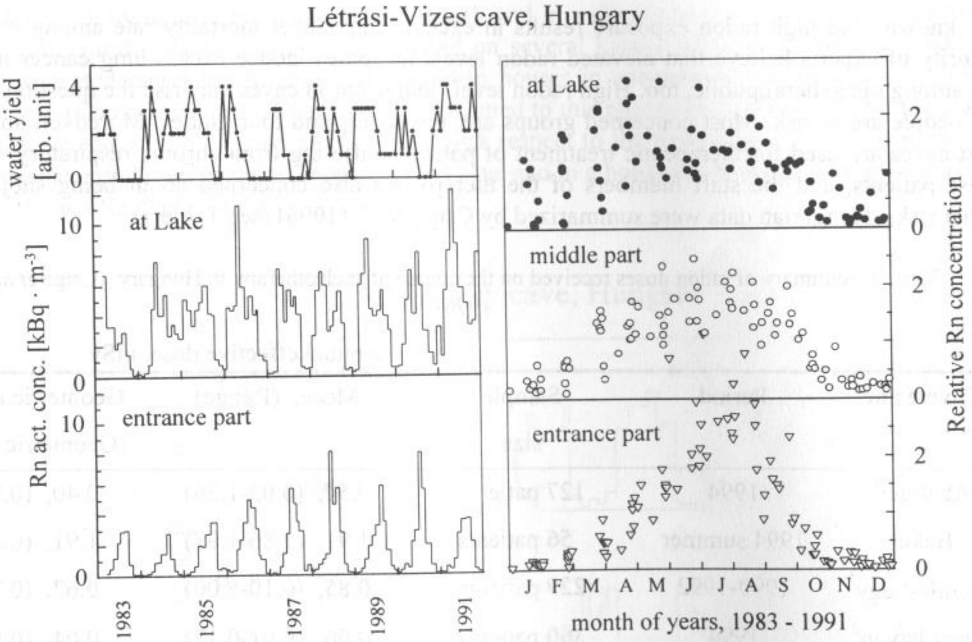

Figure 5: The yield of the stream feeding the Lake and the temporal change of radon activity concentrations in air at the Tó (Lake) and at the entrance part of the Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary (left). The form of the upper curves, in spite of low temporal resolution, shows a connection between the two quantities. The phenomenon is explained by the fact that water is the dominating radon source in cases of floods. The difference in springtime variations of normalized monthly radon distributions (right) reflects the same phenomenon, simultaneously showing radon regimes of the three distinct microclimatic zones.

The actual value of the radon concentration in the cave is influenced by subsurface fluid motions due to the periodically or randomly changing gradients in the environmental parameters (temperature, pressure, humidity, stresses,...), and by the radon concentration saturated in the pore spaces of the surrounding rocks. Using long-term temporal and large scale spatial variation measurements from the analysis of temporal radon variations, the complex interplay of the two traced substances, water and air, can be identified.

Such measurements were performed in the Létrási-Vizes cave, Bükk Mountains, Bükk National Park, NE Hungary (Lénárt *et al.*, 1988). Typical patterns of temporal changes are characterized by summer maxima and winter minima. Similar variations were found at almost all measuring sites with the exception of that part where more or less continuous water inlets were present. According to the temporal variation, Lénárt *et al.* (1988) differentiated three parts in the cave. In the first part (entrance part) the mean radon activity concentration in air increased from 1 kBqm^{-3} to 2.2 kBqm^{-3}. In the second part, it fluctuated around 2.2 kBqm^{-3}. We note that in these two parts there was a slight asymmetry in the way the radon changes between summer and winter. In the springtime, radon levels in the cave increase gradually, whereas in the fall decline rapidly. (On increasing the time resolution of the measurements, the same effect can be made much pronounced, as was found by Lively and Krafthefer (1995) using active techniques.) The highest air radon values, around 4 kBqm^{-3}, were found in the third part, which was characterized by the presence of several streams and a Lake fed by them. In the third part the dominant role of the periodic stream is evident from the long-term measurement (see Fig. 5); while in the first two parts the change of the radon activity concentration showed close connection with the temperature difference between the cave and outside.

Radon Risk Inside and Above the Caves

It is known, that high radon exposure results in excess lung cancer mortality rate among miners. A majority of experts believe that elevated radon levels in homes induce excess lung cancer mortality rate among the general public, too. High radon levels that occur in caves may rise the question whether any people are at risk. Most concerned groups are, the cavers and tour guides. Moreover, some cold karst caves are used for therapeutic treatment of patients suffering from chronic respiratory diseases. These patients, and the staff members of the therapy are also concerned about being subjected to higher risks. Hungarian data were summarized by Csige *et al.* (1996) (see Table 1).

Table 1. Summary of radon doses received on the course of speleotherapy in Hungary (Csige *et al.*, 1996).

			Annual effective dose, mSv	
Cave name	Period	Sample size	Mean, (Range)	Geometric mean, (Geometric STD)
Abaliget[1]	1994	127 patients	0.54, (0.03-1.26)	0.40, (0.77)
Béke[2]	1994 summer	56 patients	1.91, (1.86-1.97)	1.91, (0.02)
Szemlő-hegy[3]	1990-1992	229 patients	0.85, (0.10-5.00)	0.62, (0.76)
Szent István[2]	1994	360 patients	0.06, (0.01-0.17)	0.04, (0.88)
Tapolca[4]	1994	481 patients	0.87, (0.04-2.19)	0.45, (1.32)
Baradla[2]	1990-1994	12 tour guides	2.66, (0.12-5.55)	2.13, (0.80)

1 - Mecsek Mountains, S Hungary; 2 - Aggtelek Karst, Aggtelek National Park, NE Hungary; 3 - Buda Mountains, Central Hungary; 4 - Balaton Highland, W Hungary; In the case of Szemlő-hegy cave (3) cumulative effective doses are given over 1990-1992, as not all the patients attended each year to the therapeutic courses.

Currently, in Hungary radon is continuously monitored in all the five caves where therapeutic treatment of bronchial asthma and chronic bronchitis is going on. Some caves open to tourists are also monitored. In the case of the best known of these (Baradla cave, Aggtelek National Park, Hungary), the radon doses of tour guides were estimated for the years 1990-1994.

According to Navrátil *et al.* (1994) the range of patients' doses from nine different therapeutic caves in Europe is 0.07-1.32 mSv, which well fits with the above table. Based on internal working time reports, annual effective doses for therapy staff members are about 0.4 mSv in Abaliget cave, 0.12 mSv in Szent István cave and 6 mSv in Béke cave. Annual personnel exposures also cover a wide range. Tour guides in Abaliget cave receive about 12 mSv annually, Cigna and Clemente (1981) cite Yarborough, who has found 0.005-19.9 mSv per year for seven different US caves; whereas Nikodemová (1995) reports effective dose rates 0.17-4.05 mSv per month for personnel in Slovakian caves.Most active cavers may be exposed to even higher doses; so personal radon dosimetry is highly recommended for those people. One of the authors (J. Hakl) has measured his own annual effective dose due to radon inhalation while working in different caves, and found that it was higher than 30 mSv in 1992. Even higher personal doses can be achieved in caves with unusually high radon concentrations. Hyland and Gunn (1994a) estimated that it took 33 hours to reach 15 mSv limit in some caves of North Pennines in the U.K. Two important radiological consequences follow from the periodical behaviour of subsurface air circulation in karst. First is the seasonally varying underground radon level which results in a large difference between summer and winter values, and hence also in the radon exposures received by cave visiting people.

The other consequence of the seasonally directed transport phenomenon is that variation in radon exhalation can also be expected on karst terrains seasonally. A very convincing experimental result of the phenomenon is represented on Fig. 6, where radon time series measured inside the Hajnóczy cave, Hungary (with minima in the cold winter season), and in a slit above the cave (with winter maxima) are shown. Similar winter maxima were found on several karstic terrains of Hungary (Hakl *et al.*, 1992). Elevated radon levels already observed in houses in summertime (Wilson *at al.*, 1991) and wintertime (Gammage *et al.*, 1992) are both attributed to this phenomenon, showing that the season of maxima depend on the position of the house with respect to the underground air circulatory system. The inverse correlation of radon levels inside a cave and in a house above the cave was found directly by Lively and Krafhefer (1995).

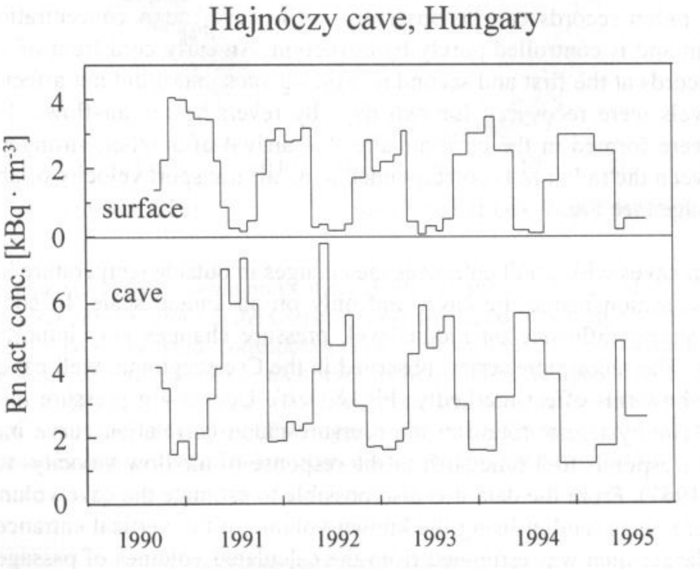

Figure 6. Push-pull type change of seasonal radon activity concentrations observed in the Nagy-terem (Great Hall) of the Hajnóczy cave, Bükk Mountains, Bükk National Park, NE Hungary, and in a surface slit above the cave.

Combination of Etched Track and Real-Time Detection Techniques

Combining etched track and real-time detection techniques, spatial and detailed temporal variation of radon concentration can be obtained. Such type of combination of continuous and integrating radon measurements was performed in the Vass Imre cave situated in the Aggtelek Karst, Aggtelek National Park, NE Hungary. It has one entrance and is situated practically horizontally with no visible vertical connection to the surface. In the middle of the cave there is a syphon, which was closed by water several times during the observation period (1987-1993). When the syphon is open, there is continuous air flow either in or out from the cave through the entrance (winter and summer, respectively). When the syphon is closed, there is no measurable air flow through the entrance. The radon concentration is being measured with etched track detectors since 1987 at 12 sites located at equal intervals along the cave passage, and in 1991 three more continuous real-time radon monitors were installed at characteristic points of the cave. Etched track detectors changed monthly showed elevated levels in the summer and lower levels in the winter. The penetrability of the syphon markedly affected the radon levels through controlling the mass of infiltrated air through the cave system. The restriction of the air flows has different effect on the forming of radon levels depending on the place of measurement (in front of *or* behind the syphon) and on the season. In summertime the restricted air flow decreases (8.5 kBqm^{-3} → 4.0 kBqm^{-3} *or* 8.5 kBqm^{-3} → 3.1 kBqm^{-3}), but in wintertime it increases (1.2 kBqm^{-3} → 2.5 kBqm^{-3} *or* 1.2 kBqm^{-3} → 3.5 kBqm^{-3}), the mean radon levels in the cave, resulting in an overall drop (4.8 kBqm^{-3} → 3.3 kBqm^{-3}) of 30% in the annual mean radon concentration . This asymmetric effect can be explained on the basis of the air circulation through covering strata (Géczy *et al.* 1988) and is in agreement with the results of numerical calculations (Holford *et al.* 1993) showing the increase of mean radon levels due to periodically changing flowing conditions. On analyzing temporal changes in radon time series, we found advection as the dominant transport process. This is also shown on the transient part of the radon record from the Vass Imre cave, Aggtelek Karst, Aggtelek National Park, NE Hungary, which corresponds to a summertime syphon opening (see Fig. 7 left). The shape of the curve, on comparing it to the results of numerical calculations on radon exhalation from porous media (Janssens *et al.* 1984), corresponds to the case when a sudden pressure drop results in instantaneous air-flow development. The pressure drop effect was also observed experimentally during a planned syphon closing and opening. On the other hand, the absence of strong daily changes in the radon record, which would correspond to the observed daily air flow fluctuations, shows the strong smoothing effect of diffusion, which can be due to relatively undeveloped fracture system of this cave. (We note that in caves located in more karstified environment, more or less pronounced daily fluctuations in radon records can be observed.) The cave radon concentration 'shut down' is very quick in autumn and is controlled purely by advection. An early cold front of short duration caused a fall in radon records at the first and second measuring sites, but it did not affected the end of the cave. High radon levels were recovered for two days by reversing the air-flow. Finally, permanent low radon values were formed in the cave air after the arrival of another, stronger cold front. The time-difference between the radon falls corresponded to an air transport velocity of about 50 m·h^{-1} along the main cave passage (see Fig. 7, right).

In some case, in caves with small entrances the changes in outside temperature have only a small effect on radon concentration inside the cave, and only on an annual scale. In contrary, the atmospheric pressure have strong influence on radon level, pressure changes play important role in controlling radon transport. The radon time series, observed in the Cserszegtomaj well-cave, Keszthely Mountain, SW Hungary, show this effect markedly (Fig. 8, left). Decreasing pressure increases the radon level and inversely. The hysteresis found in the pressure-radon correlation curve indicates nonlinearity of processes. It corresponds to a time shift in the response of air-flow velocity to the pressure changes (Wigley *et al.* 1967). From the data it is also possible to estimate the cave volume, using simultaneous pressure measurements (and utilising the known volume of the vertical entrance well). We found it to be 8-10 times larger then was estimated from the calculated volumes of passages. On the right part of the figure, for the sake of comparison, also displayed the radon response of a chimney effect controlled cave system.

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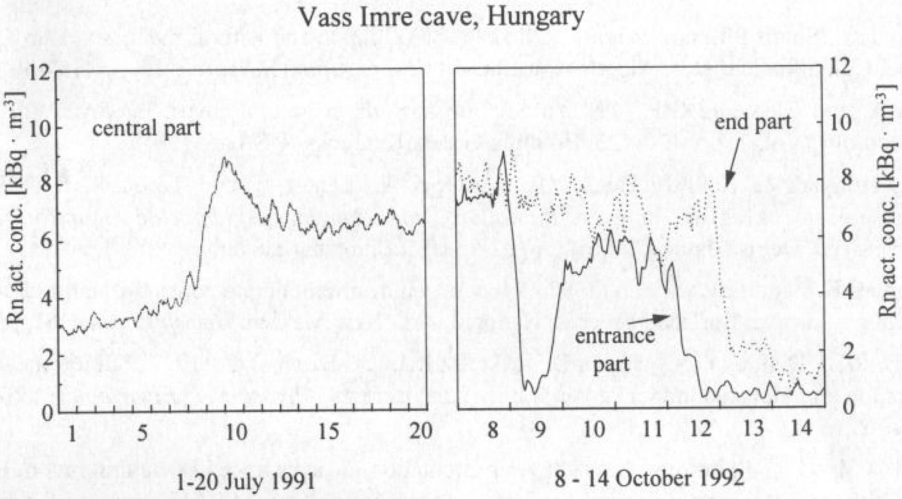

Figure 7: The real time records of radon time series from Vass Imre cave, Aggtelek Karst, Aggtelek National Park, NE Hungary, at the times of syphon opening (left) and arriving of early cold fronts during fall (right).

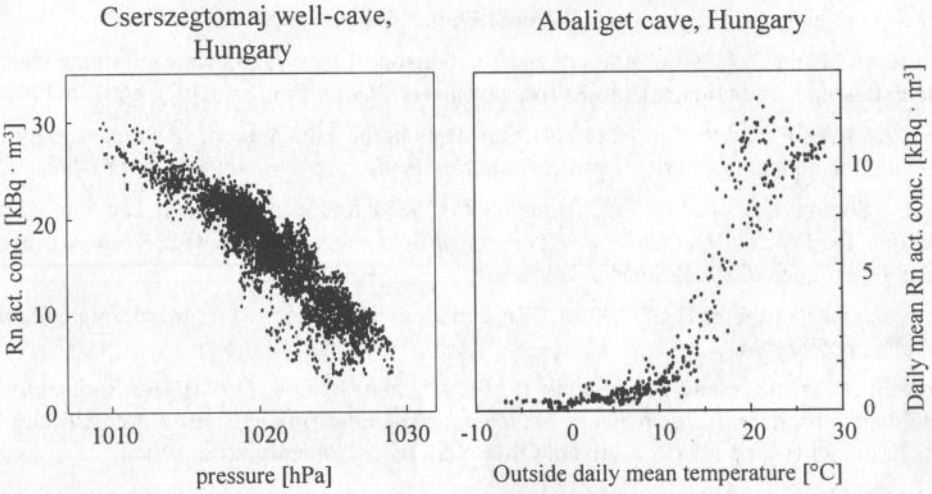

Figure 8. Radon activity concentration vs atmospheric pressure observed at one of the five continuous observation stations in the Cserszegtomaj well-cave, Keszthely mountains, SW Hungary (left). The 'data cloud' corresponds to the 2 month section of the continuous data record. The inverse pressure control on radon levels is well seen in the figure. On the right-hand part of the figure we have displayed radon levels vs outside temperature observed in the Abaliget cave, Mecsek mountains, S Hungary. The S-type dependence of radon concentration on the outside temperature is typical for caves dominated by chimney effect winds.

ACKNOWLEDGMENT

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④

APPLICATION OF RADON-222, AS A NATURAL TRACER IN ENVIRONMENTAL STUDIES

Ph.D. thesis

Dr. József Hakl

Lajos Kossuth University
Debrecen, 1997

Ezen értekezést a KLTE **fizika** doktori program *fizikai módszerek alkalmazása interdiszciplináris kutatásokban* alprogramja keretében készítettem 1995 - 1997 között és ezúton benyújtom a KLTE doktori Ph.D. fokozatának elnyerése céljából.

Debrecen, 1997 szeptember

Dr. Hakl József jelölt

Tanúsítom, hogy Dr. Hakl József doktorjelölt 1995 - 1997 között a fent megnevezett doktori alprogram keretében irányításommal végezte munkáját. Az értekezésben foglaltak a jelölt önálló munkáján alapulnak, az eredményekhez önálló alkotó tevékenységével meghatározóan hozzájárult. Az értekezés elfogadását javaslom.

Debrecen, 1997 szeptember

Hunyadi Ilona

Dr. Hunyadi Ilona témavezető

Contents

Preface

Copy of relevant publications

Preface

It happened in October 1985 that Dr. György Somogyi, the late leader of the Track Detector Group of the Institute of Nuclear Research of the Hungarian Academy of Sciences (Debrecen, Hungary) invited me to accompany him on one of his favourite duties. He took me to a cave (Hajnóczy cave, Bükk Mountains, Hungary) not for recreation but to work, to change the etched track type radon concentration measuring devices. It was when I have got my first impression about the application of a nuclear technique in environmental studies. The underground tour lasted 6 hours. I got stiffness for two days and I promised myself never to repeat this action again.

It did not happen though. The things around caves started to interest me and I readily took part in field-work. In the spring of 1987 Dr. György Somogyi suddenly died of heart attack. Dr. Ilona Hunyadi took the lead of the group and under her leadership I got the chance to continue the work in this field. I have elaborated the available cave radon data and started to organise the continuation of cave studies. It has became and still it is my main scientific area. I have enjoyed this work very much.

It was Dr. Ilona Hunyadi, who in the early hard times fought with great enthusiasm and success for financial background of radon works; exposing her young colleagues, in the meantime, to the national and the international scientific community. Here I should like to thank for her selfless help, advice and all she had done for the group.

Also, I should like to thank my closest colleagues Dr. István Csige and Attila Vásárhelyi, with whom I had the chance to work together in the laboratory.

During the field and laboratory works my closest colleagues were Gábor Géczy, Dr. András Várhegyi, Dr. János Somlai, Dr. László Lénárt, Dr. István Törőcsik, Dr. J. L. Seidel, Ferenc Szolga and László Rénes, with whom I spent exciting times discovering the secrets of the nature.

Particularly, I should like to thank to Mrs. Enikő Molnár for the excellent technical and administrative work and for all she has done for us in the Radon laboratory of Debrecen.

During the course of the last ten years these radon studies were financed by the Hungarian Academy of Sciences Research Fund contract No. 1-3-86-185 and Hungarian National Scientific Research Fund contract Nos. 2011, 3005, T 016558, T 017560. Their support is highly acknowledged.

1 Introduction

With increasing pollution world-wide the need for understanding of the dynamics of environmental processes has became of primary concern of mankind. The problems are focused on two, in certain sense opposite areas. These are the industrially heavily contaminated areas and yet clean but endangered regions. In the first area questions regarding the propagation of contaminants, in the second one the transport processes themselves are in the centre of public and scientific attention, respectively.

One of the methods suitable to study these processes, which reflect the interaction of atmosphere, hydrosphere and lithosphere, is the use of tracer isotopes. Subsurface natural fluids in the majority of cases carry small amounts of radioactive isotopes. Among them the alpha radioactive ^{222}Rn, as a member of ^{238}U decay series, is ubiquitously present in the ground and in the lower atmosphere as well. Through the measurement of radon concentration in the geological environment, information can be obtained about the transport processes as well as about penetrated geological structure. This approach is applicable also in cases of radioactive contaminants, where the essential question is the retention of transport.

Among the endangered regions in highly permeable areas, as e.g. karst[1] is *per se*, the problems are perhaps the sharpest, as they serve as natural water reservoirs for mankind. In these areas, owing to the set of interconnected fractures and voids, the propagation of external effects is very quick and deep. One of the most appropriate methods to trace transport with natural radon in these systems is the application of etched track detectors allowing desirable large-scale *in situ* measurements. At the Institute of Nuclear Research of the Hungarian Academy of Sciences first underground environmental alpha radioactivity studies, based on the latter technique, were initiated by Dr. G. Somogyi in 1977. I have entered this step by step widening radon field in 1987. My scientific interest focused on studying radon transport, giving emphasis to cave investigations.

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[1] Areas in which the rock has suffered solutional activity with the development of caves and enlarged fractures

Caves occur everywhere on the earth, although mainly in karst areas - as they are formed mostly in limestone environments. They may appear to men as static environment, where not only the continual darkness but also the temperature and high relative humidity is stable. This belief is, however, not justified, because fluid currents can cause measurable changes in the physical parameters. The underground radon measurements supplied us with a great amount of data, both spatially and temporally variable, confirming the awaited diversity of data. The complexity of environmental transport processes manifests itself everywhere. The influence of inflowing waters, the presence of underground air circulation as well as the morphology and structural dependence of transport processes are traceable in our radon records. Based on these investigations, physical model interpretations of the observed fluid motions were developed in collaboration with the Department of Physical Geography, Eötvös Lóránd University (Budapest, Hungary) and Department of Hydro- and Engineering Geology, Miskolc University (Miskolc, Hungary). The so-called vertical geogas microbubble radon transport model was developed in collaboration with Mecsek Ore Mining Ltd (Pécs, Hungary). Its validity conditions were tested in measurements done in a 270 m deep karst well at Miskolc University and on a 8.5 m high model column at the Laboratory of Hydrogeology, University of Montpellier (Montpellier, France). These collaborations resulted in developing of a microprocessor controlled automatic radon-measuring unit in Hungary, which, since 1991, is routinely used in fieldwork parallel with track etch technique.

The object of this thesis is to summarise results obtained during the last decade. I have included those findings, in obtaining of which my role was decisive. As a main part, I will outline results related to radon concentration measurements in karst caves and in natural (not necessarily karst) waters. Additionally, I will summarise results of methodological developments connected to the solutions of tasks of environmental radon activity concentration measurements. These are the studies of radon transport through different filter and blocking materials, and the developments of new measuring techniques for underwater and continuous radon concentration measurement purposes.

2 Review of literature

2.1 Sources of radon in karst

Radon is a mobile, chemically inert radioactive element. All the three naturally produced isotopes, ^{222}Rn (radon), ^{220}Rn (thoron), and ^{219}Rn (actinon) decay by emitting alpha particles. These noble gas isotopes are produced from radium decay as steps in lengthy sequences which originate from uranium or thorium series – ^{222}Rn from ^{238}U; ^{220}Rn from ^{232}Th; and ^{219}Rn from ^{235}U. Their respective half lives are 3.82 d, 55.6 s, and 3.96 s (mean lives 5.51 d, 80.2 s, and 5.71s). The relative importance of the three isotopes increases with their mean lives and relative abundance. ^{219}Rn is the shortest lived, and is virtually always produced in much smaller amounts than is ^{222}Rn, since the natural ^{235}U/^{238}U ratio of these ultimate progenitors is 0.00719. Hence ^{219}Rn is largely ignored. ^{220}Rn too is short lived relative to ^{222}Rn and consequently moves a much smaller distance from its source than does ^{222}Rn. In air, for diffusion constant, D, of 0.1 $cm^2 \cdot s^{-1}$, the mean distances of diffusive motion are 2.2 m for ^{222}Rn and 0.029 m for ^{220}Rn. Hence in circumstances where signals from relative distant sources or processes in the earth are sought, ^{222}Rn is by far the dominant nuclide, and ^{220}Rn provides only a local background that one want to exclude during detection. Characteristic feature of radon isotopes is their high mobility in comparing them to other members of the radioactive decay series. They are able in a short time to escape into the pore space from the mineral in which they are born. The radon atom that escapes is either released by direct ejection by recoil from alpha emission (Kigoshi, 1971) or by diffusion through damaged channel after chemical solution of it with pore water (Fleischer and Raabe, 1978). From the pore space radon atoms migrate towards microcracks, fractures and cave volumes either by diffusion or forced flow.

The sources of radon in caves are the bedrock and deposits. Radon levels in caves are determined primarily by the uranium content of the rock. Limestone and other sedimentary rocks are found to contain about 1.3 - 2.5 ppm ^{238}U (16 - 31 $Bq \cdot kg^{-1}$) on average. The relatively high values of radon found in caves are due to these minute quantities of parent substances that occur naturally on and within the interior surfaces of the caves. Increased ^{238}U concentrations can be associated either with fluorite mineralizations or hydrocarbons present in the surrounding limestone.

Uranium can be oxidised and mobilised by groundwater flow. Once reducing conditions are encountered, the uranium is readily precipitated from solution. This leaching fixation process leads to the enrichment of uranium in adjacent deposits. This secondary transport and enrichment process is important in cave environment, as fractures in rock can increase the surface area interacting with water. Experimental evidence of this effect in cave environment was found by Navrátil *et al.* (1993), who observed enrichment of uranium on cave walls.

The immediate radon source in caves is the ^{226}Ra content of the rock. Nazaroff *et al.* (1988) reports mean ^{226}Ra content of carbonates to be 25 $Bq \cdot kg^{-1}$ (range 0.4-233), which were found to be distributed lognormally. The results of ^{226}Ra determinations of bedrock and soil samples from the Hungarian caves examined fall in range 0.6-26 $Bq \cdot kg^{-1}$ (Hunyadi *et al.*, 1997). The relatively impermeable soils (deposits), such as clay, do not have sufficient porosity to allow transfer of significant amounts of soil gas, therefore their contribution to radon budget is small (Michel, 1987). Accordingly, Burkett (1993) found radon emitted from the clay to be not sufficient to account for the radon concentrations measured in the cave.

2.2 Radon as a tracer

Subsurface natural fluids in the majority of cases carry small amounts of environmental isotopes. The behaviour of these elements, and the variation of their concentration in time and space, is the result of physical, chemical and biological interactions. These elements, as their physical properties, concentrations, etc., provide information on flow and kinetics of the carrying substances, are called natural tracers.

The application of radon as a natural tracer is not yet common and widespread. Among the natural tracers it would be considered on the one hand as ideal since it is easily detectable even in small quantities, which do not modify the characteristic of the environment. On the other hand, unfortunately, its sources appear everywhere and are spread over in a manner unknown *a priory*. Therefore, the interpretation of the

concentration data is not straightforward: it needs interdisciplinary expertise of hydrogeologists, geologists, physicist, radiogeochemists, etc. Joint efforts have given results in different fields. The observations of subsurface fluid motions traced by natural radon were followed by new ideas about the basic transport phenomena and, later, by new interdisciplinary applications: as, for example, mapping of active faults; investigations of volcanic and seismic activities; earthquake prediction; hydrogeological research, etc. (Fleischer, 1988).

In the speleology, similarly to the previously mentioned fields, these types of measurements have already found their applications, and they give important contributions to the better understanding of the natural regimes of caves. Cunningham and LaRock (1991) delineated six microclimate zones in Lechuguilla cave, Carlsbad caverns, National Park, New Mexico using radon grab sampling in conjunction with observed airflow data. Atkinson *et al.* (1983) from a single set of etched track measurements in the Castleguard cave, Columbia icefields, Alberta, Canada, identified the effect of tributary air currents from larger fissures.

The most common and most apparent phenomenon, which was discovered in the majority of the investigated caves throughout the world, was the annual change of radon activity concentration. Wilkening and Watkins (1976) identified temperature gradients favourable to vertical convective transport through relatively large openings. They identified as well transport of radon by air movement through cracks and fissures due to pressure gradients (Wilkening, 1980). As karst caves are situated generally in highly fracturized rocks, such a configuration is favourable for the emergence of air circulation through this fracture system. The strength of such air motions is taken to be proportional, to a first approximation, to $\Delta T/f$, where ΔT is the temperature difference between the cave and outside and f is a friction factor characterising the flow resistance (Wilkening and Watkins, 1976; Atkinson *et al.*, 1983; Quinn, 1988).

2.3 Modelling of Observed Features

The underground radon transport can be described by the following transport equation:

$$\frac{\partial C}{\partial t} = D_{eff}\Delta C - \nabla(\vec{v}C) - \lambda C + \phi,$$

where C $[m^{-3}]$ is the radon concentration in pore space, D_{eff} $[m^2 \cdot s^{-1}]$ is the effective diffusion coefficient of radon, $\vec{v}\,[m \cdot s^{-1}]$ is the velocity of the carrying substance, λ $[s^{-1}]$ is the decay constant of radon and ϕ $[m^{-3} \cdot s^{-1}]$ is the source term. In the equation first term describes diffusion, second term advection, third term decay and fourth term sources of radon. For the solution of transport equation, first, it is necessary to know the velocity field (e.g. Navier-Stokes equation, which by itself is a sufficiently complicated problem); then taking into account the source term, and the initial and boundary conditions C can be determined. The emerging phenomena are determined by the form, shape and structure of the underground void space. The above equation generally can be solved only numerically.

The realisation of radon transport processes sharply depends on the configuration of the interconnected underground cavities. In the case of blind end systems, atmospheric pressure variation are the main control parameter (Wigley, 1967; Ahlstrand, 1980), which are superimposed by convective air exchange due to temperature differences in cave systems with vertical extension. In the case of relatively large entrances, the convective air exchange due to temperature differences can mostly account for the radon transport process taking place (Wilkening, 1979). In the case of two or more entrance systems, where the other 'entrances' can be complexes of smaller fissures and fractures, chimney effect winds may dominantly govern the radon transport, or in some cases atmospheric winds may do so.

The interpretation of the data can be affected by the presence of these unknown (unassumed) 'entrances'. Yarborough (1980), in a study of nine caves, identified two general types of physical cave configurations that affect airflow patterns and radon concentrations. Type 1 caves have most passages at or above the entrance elevation, Type 2 caves have most passages below the entrance elevation. When the outside temperature exceeded the cave temperature, he found that Type 1 caves exhaled, while stagnation occurred in Type 2 caves: when the outside temperature fell below the cave temperature, both caves inhaled. As Type 1 cave is horizontal "mirroring" of Type 2 cave the seasonal ventilation patterns should be alternatives of each other. The clear asymmetry in this case, however, points to the difference between the air flow-through and blind end system. This indicates, that substantial hidden parts of the upward directed systems remained unrevealed from the descriptive point of view.

The above examples illustrate the problems of interpretation and modelling. In two special cases, however, the emerging processes are analytically easy to survey. These are the cases of idealised two-entrance horizontal flow-through (type A), and one narrow entrance vertical caves (type B). In the case of the horizontal model cave, the second entrance may represent the set of vertical fractures, which connect the main passage to the surface through the overburden.

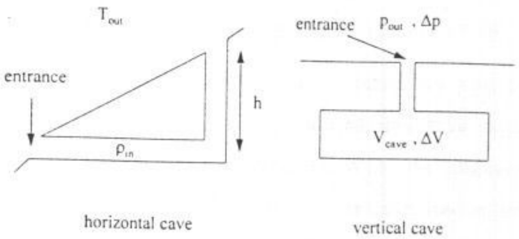

Figure 1: Schematic horizontal (type A) and vertical (type B) model caves; h is the elevation difference between the upper and lower entrances (horizontal cave), ρ_{in} is the air density of inside air, T_{out} is the outside temperature, p_{out} is the ambient outside pressure, V_{cave} is the cave volume and ΔV is the volume of air passing the vertical cave entrance in case of Δp change in the ambient outside pressure.

First, let us consider a schematic horizontal cave (see Fig. 1, left). In this case, owing to the temperature dependence of air density the pressure exerted at the lower end of the cave by the outside air will differ from the inside one. This pressure difference in this case is (Atkinson *et al.*, 1983):

$$\Delta p \approx -gh\rho_{in}\frac{\Delta T}{T_{out}},$$

where ρ_{in} is the density of inside air, g is the gravitational acceleration, h is the height difference between the two entrances, ΔT is the temperature difference between the cave and the outside, and T_{out} is the outside temperature. According to this relation, the direction of airflow through the entrance depends on the season; in warm season air flows out of, and in cold season air flows into, the cave at the lower entrance.

A more realistic model of fracturized karstic overburden above a horizontal cave is the model of parallel vertical voids connecting the main passage to the surface (Scheidegger and Liao, 1972). The variation of ^{222}Rn activity concentrations in this case is interpreted by an air circulation model (Géczy *et al.*, 1988). According to it, in warm season, air will flow into the cave from the direction of the radon-rich fracture system (and will flow out of the cave through the entrance at lower altitude) resulting in high radon levels in cave. In cold season, the direction of the flow is reversed. From the direction of the lower entrance fresh outside air flows into the cave forming considerably lower radon concentration levels than in summer

In the case of narrow-entrance vertical caves, the most effective processes in inducing air motions are the atmospheric pressure changes (Fig. 1, right). Falling ambient atmospheric pressure drains air from the cave; increasing atmospheric pressure presses air into the cave through the entrance. The volume of air passing the cave entrance:

$$\Delta V \approx -\frac{V_{cave}}{p_{out}}\Delta p\,,$$

where V_{cave} is the cave volume, p_{out} and Δp are the ambient atmospheric pressure and the change in the ambient atmospheric pressure, respectively. Airflows induced by atmospheric pressure changes can be rather quick. At the entrances of giant caves their speed can reach several tens of km·h^{-1}(Cunningham and LaRock, 1991).

3 Materials and methods

For the purpose of environmental radon concentration measurements opened diffusion chambers equipped with LR-115 type II alpha sensitive polymer track detector were used. The diffusion chamber consisted of a cup (diameter: 5.5 cm, height 12 cm) with a detector located at the bottom of the cup. The other end of the diffusion cup was opened to the atmosphere and looked downwards during the exposure. The discrimination against thoron (^{220}Rn) was obtained by the delay effect of the diffusion on the basis of the use of a sufficiently long cup. The sensitivity of the detector is 2.3 alpha-tracks·cm^{-2}/kBq·m^{-3}·h at standard etching conditions (2.5 N NaOH, 60 °C, 2.5 hours). Generally, in each cave the track etch detectors were placed along the main passages of caves more or less equidistantly. There were generally 3-20 regular measuring sites per cave. Extra observation points were established at "characteristic" places. Typical exposure time was 1 month. About 10000 observation data were obtained in 31 investigated Hungarian caves during the years 1977-1997.

From the beginning of the 1990s, microprocessor controlled automatic field radon monitors (type Dataqua[#]) were gradually introduced into cave studies. The single channel type measured only radon concentration, while the multiparameter version simultaneously registrated the temperature and air pressure. Radon concentration was measured in 1 hour cycles using open type diffusion chamber (delay time ≈ 1000 s) equipped with alpha sensitive Si based semiconductor detectors. The sensitivities of the units are 6.7 cph/kBq·m^{-3}·h (detector sensitive area: 1 cm^2) and 17.8 cph/kBq·m^{-3}·h (detector sensitive area: 3 cm^2) with an 0.1 cph initial background (cph stands for count per hour). Generally, in each cave the continuous radon monitors were placed in the main passages at characteristic places. There were 1-5 regular measuring sites per cave. A total number of approximately 400000 hourly readings was obtained with 11 monitors in the investigated five caves during the years of 1991-96.

[#] Produced by Dataqua Ltd., Kölcsey F. u. 1, 8220 Balatonalmádi, Hungary

Measuring sites

The most characteristic results were obtained in the following caves from the investigated ones:

Bükk Mountains, Bükk National Park, Hungary

- The *Létrási-Vizes cave* is a multi-level typical swallet cave. The main passage declines towards the end of the cave, which is located at 85 m depth from the natural entrance. The cave collects waters from the surrounding area, but its streamlets often go dry. The cave can be considered as cave of type B. The radon concentration was measured with etched track detectors at 15 places along the main passage during the period of 1983-1993.
- The *Hajnóczy cave* can be considered as cave of type A. Its passages are located along 3 parallel fault zones. The distance between two farthest points of the cave is 150 m. The radon concentration was measured with etched track detectors at 8 places during the period of 1977-1982, at 6 places in period of 1982-90, at 9 places since 1991; and with one Dataqua radon monitor since 1991 in the central fault zone.
- The *Szepessy cave* is a cave of type B. The depth of the entrance shaft is 130 m, the maximum depth of the cave is 165 m. The radon concentration was measured with etched track detectors at 4 places during the period of 1983-1985 and at 9 places in 1991-1992.
- The *Istvánlápa cave* is a cave of type B. It is one of the deepest caves in Hungary. The depth of the entrance shaft is 210 m, the maximum depth of the cave is 240 m. The radon concentration was measured with etched track detectors at two places, in 130 m and 215 m depths in years 1983-1985 and with two Dataqua radon monitors in summer 1994.

Aggtelek Karst, Aggtelek National Park, Hungary

- The *Baradla cave* is nearly a horizontal cave located between Aggtelek and Jósvafő villages. The cave has several entrances located along and at the ends of the main passage, which length is 6 km. The main passage declines towards the Jósvafő entrance. The elevation difference between the entrances at Aggtelek and Jósvafő is 60 m. The radon concentration was measured with etched track detectors at 11 places in 1984-1986, at 13 places in 1987-1995 and with one Dataqua radon monitor in 1991-1997.

- The *Vass Imre cave* is a cave very similar to the model cave of type A. It has one entrance and the main passage is situated horizontally. The length of the cave is 600 m. In the middle part of the cave there is a siphon, which was closed by water two times during the overall observation period. The radon concentration was measured with etched track detectors at 5 places in 1980-1985 and at 15 sites in 1987-1994. Additional three Dataqua radon monitors were operated in the cave in the period 1991-1997, one at the entrance, one in the middle and one at the end of the main passage.

Pilis Mountains, Hungary

- The *Sátorkő-puszta cave* is a vertical cave of type B. The depth of the cave is 48 m. The radon concentration has been measured with etched track detectors at 2 places, in 33 m and 45 m depths since 1991.

Mecsek Mountains, Hungary

- The *Abaliget cave* is very similar to the model cave of type A with a small flowing stream inside the cave. It has one entrance and the main passage is situated practically horizontally. The length of the main passage is 500 m. The radon concentration was measured with etched track detectors at two places in 1980-1984. Additionally we operate three Dataqua radon monitors in the cave since 1991, one at the entrance, one in the middle and one at the end of the main passage.

Buda Mountains, Hungary

- The *Szemlő-hegy* cave is a horizontal cave with the main entrance situated at lower altitude, and several other 'entrances' located at higher altitudes of the hill. The cave passageways are situated in two levels but the cave can be considered as cave of type A with one dominating large opening to the surface. The length of the lower passage is 350 m. The end of the main passage is 65 m depth from the surface, the vertical distance between two levels is 15 m. The radon concentration has been measured at 10 places since 1985 and with two Dataqua radon monitors at both levels of the cave since 1992.

Keszthely Mountains, Hungary

- The *Cserszegtomaj well cave* is a horizontal maze cave of type B, which was formed in 50 m depth on the boundary of dolomite and sandstone. It has one 50 m deep artificial vertical entrance. The whole formation is covered by clay. The radon concentration is unusually high due to the sandstone environment of high porosity and bad ventilation. The radon concentration was measured with etched track detectors in 1984-1986 and at 10 places in 1992-1997. We operated 5 Dataqua radon monitors in the cave during the period of 1993-1996, one at the bottom of the entrance, other four in the depth of the cave.

Lamalou Karst, France

- The *Lamalou cave* is a horizontal water cave with a flowing stream inside the cave. A part of the cave, except floods, is aerated. The aerated part of the cave has one vertical and one horizontal entrance. The aerated part of the cave system can be considered as cave of type A. The length of the vertical entrance shaft is 20 m, the length of the horizontal part of the aerated passage is 90 m. During the period of 1991-1997 we operated 2 Dataqua radon monitors at the 15 m depth, one in the vertical entrance shaft and one on the roof of the horizontal passage deeper inside the cave.

4 Summary of results

(Papers referred as [A...] containing the new scientific results can be found in the attachment.)

1. Characterisation of airborne ^{222}Rn concentration occurrences in karst caves

1.a Compiling all the globally available radon concentration data from 220 different caves world-wide I have found, that the distribution of ^{222}Rn concentrations is lognormal [A1, A2] (GM=1130 Bq·m^{-3}, GSD=6.3), which is in good accordance with the awaited distribution for a geochemical element. The lower end of the scale is associated either with big cave chamber volumes or high ventilation rates; the upper end is characterised by closed, badly ventilated places and uranium-rich sediments.

1.b For the 31 examined Hungarian caves I have determined the cave average annual mean radon activity concentrations, 0.3-20 kBq·m^{-3}, the characteristic annual maximum/minimum ratios, 2-50, and the periodicity, which was typically one or half a year [A3]. In the majority of cases the marked seasonal variations can be characterised with high radon concentration values in summer and low radon concentration values in winter. In few cases, reversibly, summer minima and winter maxima were observed.

1.c I have observed a long-term variation of the annual mean radon activity concentration in all the studied caves [A1, A4]. The phenomenon shows the effect of slowly changing environmental parameters on radon transport processes. Such an environmental parameter may be the annual precipitation. The variation of annual precipitation, which may due to climatic changes, influences water content of the cave embedding rocks, which on the other hand affects radon emanation power of rocks. The long-term variation can be amplified selectively at different sites in a given cave. This latter effect is markedly shown on time series taken at two different depths of the Sátorkő-puszta cave. The ratio of the difference of data pairs to the mean of the same data pairs increases with years (see Fig. 4. in [A1]).

2. Influence of karstification and morphology of caves on airborne ^{222}Rn concentrations

2.a By model calculations I have shown, that the saturation value of radon activity concentration in fractures and the radon exhalation from fractures strongly depend on the size of the fracture. According to calculations, at low airflow velocity to aperture size ratios, the advective ^{222}Rn transport along a fracture is strongly reduced by lateral diffusion inside the fracture [A1, A6]. These theoretical predictions were justified by measurements done in the Vass Imre and Lamalou caves. In the Vass Imre cave, which can be characterised by relatively undeveloped fractures with small size openings, I found no daily variation in the radon records [A1], in spite of the observed daily air flow variation. On the other hand, in the Lamalou cave, which is embedded in a well karstified strata, characterised by big solutional openings and fracturized volumes, strong daily fluctuations of ^{222}Rn activity concentrations were recorded [A7].

2.b I have found that in horizontal caves the number of vertical fracture systems communicating the surface affects the width of the outside temperature interval, which characterises the transition from the low to the high daily average radon concentration values [8]. While in the Vass Imre cave the transition width is around 5 °C, it is around 10 °C in the Abaliget cave. In the Vass Imre cave, only one less developed fracture system, in the Abaliget cave, more than one more-developed vertical fracture systems exist. The step by step widening of the transition interval as a function of the number and size of openings, is further supported by the data obtained in the Szemlő-hegy cave. Here the step function type curve is practically reshaped to a linear function (see Fig. 3. in [A8]).

3. Identification of air circulation pathways and microclimate zones in cave systems based on airborne ^{222}Rn concentration measurements

3.a I have pointed out, that in a consequence of the seasonally directed underground transport, enhanced surface exhalation can be expected on karstic terrains seasonally [A3]. Experimentally the existence of the latter phenomenon was verified in the Bükk Mountains, Hungary, where the radon time series measured inside the Hajnóczy cave and in a slit above the Hajnóczy cave were inversely correlated (see Fig. 6. in [A2]) showing high winter radon concentration values on the surface. Winter maxima were as well observed on several karstic terrains of Hungary [A3]. The above phenomenon can serve as an explanation to other observations published in the literature.

3.b I have identified microclimate zones based on the analysis of temporal variations of radon concentrations in caves. In the Baradla cave these zones are [A5]: 1. the entrance region in the Aggtelek part with summer maxima and winter minima; 2. the middle part from Libanon hill to Vöröstó entrance with constant radon levels; and 3. the region between the entrances of Vöröstó and Jósvafő with winter maxima and summer minima. I have pointed out, that the temporal behaviour of radon activity concentration in the third microclimate zone of the Baradla cave refer to a definite connection between the main passage of the Baradla cave and the Rövid-Alsó cave situated at a lower altitude. Based on the difference in temporal variation of radon concentrations three microclimate zones were as well identified in the Létrási-Vizes cave [A2, A9].

4. Determination of underground airflow velocity and its relation to cave average annual mean airborne ^{222}Rn level

4.a I have determined the propagation speed of outside fresh air intrusions underground and a volume of a vertical cave from the continuous measurements of radon concentration [A1, A2]. In horizontal caves fresh air intrusion appears when the outside temperature falls below the cave air temperature, while in vertical caves their presence is related to an increase of the ambient outside pressure. Utilising the time differences among radon falls measured at distinct places, their propagation speed can be calculated. This speed is about 50 $m\cdot h^{-1}$ along the main cave passage of the horizontal Vass Imre cave. As the length of the cave is 600 m, the whole known volume of the cave is flushed through with outside air in 12 hours. Contrary, the propagation speed of the dilution effect in the vertical Cserszegtomaj well cave is around only 2 $m\cdot h^{-1}$ in a 10-20 m region from the bottom of the entrance well. This low value shows that the cave is highly unventilated. In average 2 hPa of atmospheric pressure increase flushes out radon gas from the complete volume of the vertical entrance. Taking into account the known volume of the entrance well and the ideal gas law, it indicates, that the volume of the cave is in the order of 10000 m^3, an order of magnitude larger, than it was estimated from the volumes of known passages.

4.b I have found that cave average annual mean airborne ^{222}Rn levels depend on the strength of underground air motions [A2, A8]. In the Vass Imre cave the volume of infiltrated air is naturally controlled by the penetrability of the siphon. When the siphon is open, there is continuous air flow either in or out from the cave through the entrance (winter and summer, respectively). When the siphon is closed, there is no measurable air flow through the entrance. The restriction of the air flows resulted in an overall drop of 30% in the annual mean radon concentration level in the cave, accompanied by the decrease of the amplitude of the seasonal radon concentration variation. This effect can be explained on the basis of the air circulation through the cave covering strata. It shows the increase of mean radon levels due to periodically changing flowing conditions. The absence of seasonal variation in radon levels and the low radon activity concentration values found in the deepest parts of the narrowest vertical caves in Hungary (Szepessy, Istvánlápa) may also be attributed to this phenomenon [A3], as from the point of view of ventilation the deep caves can be considered as the most closed ones.

5. ^{222}Rn transport in water

5.a I have found that water inlets can be significant sources of radon in cave. In deeper parts of the Létrási-Vizes cave the regular seasonal variation of radon levels are considerably disturbed [A1, A2, A4, A9]. Approaching the endpoint of the cave, the periodical character of the readings decreases and the variation of springtime values becomes higher. On the other hand, there is high similarity between temporal variations of the yield of one of the intermittent streams and radon concentration measured in the air. As subsurface waters permeating porous rocks can be significantly enriched in solved radon, this effect is explained on the way that radon is essentially carried to the place by the latter intermittent stream.

5.b Using radon as a tracer I have found that thermal gradient induced convectional mixing is taking place in the water column of a 270 m deep karst well [A4, A10]. I have developed a model to calculate the depth dependence of the vertical transport velocity from the measured vertical ^{222}Rn concentration profiles. The results indicated continuous upward transport of radon in the water column with a mean velocity of about 0.7 m·h^{-1}. I have pointed out, that this high value can be attributed to the vertical thermal gradient induced convectional mixing of water, and I rejected the possibility of transport by geogas microbubble theory. The strong effect of vertical mixing on transporting radon was observed as well in model experiments at the Hydrogeological Department, University of Montpellier, France, in an 8.5 m high model column [A11]. As the search for new U ore deposits in Hungary in late 80s was partly based on the interpretation of the vertical radon profiles taken underwater in shallow drills, the recognition of underwater vertical mixing processes along bore holes in this respect played a supplementary role.

6. Methodological developments

6.a Silicon photodiode based Dataqua radon monitoring devices were developed and brought to successful applications with my significant contribution [A12]. The idea of utilising silicon detectors for field radon measurements (by alpha particle detection) came from our French collaboration partner, Hydrogeological Department, University of Montpellier, Montpellier, France. The hardware of the device (analog and digital electronics) was developed by the Dataqua Electronic Ltd. The design of the sensitive volume of the radon head of the instrument and the electronic testing were done by me at the Institute of Nuclear Research. I have calibrated the instrument in the radon reference chamber of the Swedish National Institute of Radiation Protection (Stockholm). The obtained calibration coefficient, 6.7 cph/kBq·m^{-3}·h for the 1 cm^2 surface detector, coincides well with that I have previously calculated.

6.b A novel method to measure radon permeability of thin foils using etched track technique was developed with my significant contribution [A13]. During the measurement the sample foil separates the radon source volume from the measuring volume. I have elaborated an analytical model to describe the non steady state temporal variation of radon activity concentrations in source and in the measuring volumes as a function of the permeation characteristic of the investigated foil. This method allows to determine radon permeability coefficient as well as radon/thoron separation factor of a given foil in few hours. I have tested the reliability of the method in a series of laboratory measurements using polyethylene foils of different thickness. The obtained permeability value, $7.1*10^{-8}$ $cm^2 \cdot s^{-1}$, is in good agreement with the literature data. The permeation characteristics of different materials are of interest from the point of view of sealing homes against invasion by radon. As a practical application of the method we have examined six types of floor covering materials, and found, that they can reduce radon entry into dwellings by 2-3 orders of magnitude.

6.c Two experimental techniques for the determination of the effective diffusion coefficients of radon in polymer/silicate gels and clay suspensions were developed with my significant contribution [A16, A17]. Similarly to the previous method, one side of the samples was exposed to the radon source, the other side was closed by a small measuring volume. Track etch type radon monitors were used to measure the radon exposures on both sides of the samples. The diffusion mass transport in the sample was numerically modelled for different exposure times and sample thicknesses. Diffusion constant was a fitting parameter to obtain best fit to the experimentally measured radon exposures. I used Dataqua type continuos radon monitors to test the experimental arrangement against leaks. The procedure was based on testing of the variation of the diffusion constant in subintervals of the full exposure time. The variation in the value of the diffusion constant was the indicator of leakage. We have determined the effective diffusion coefficient of radon in polymer/silicate gel-containing porous media, $3.3*10^{-6}$ $cm^2 \cdot s^{-1}$, and in Montax/clay suspension, $6.0*10^{-6}$ $cm^2 \cdot s^{-1}$. The corresponding values for the bulk phases are comparable to that characteristic in pure aqueous solutions, therefore the sealing technology applying these materials can be very attractive.

6.d A new method for the determination of the radium and dissolved radon content of water samples using track etch type radon monitors was developed with my significant contribution [A14]. During the measurement an immersed small volume radon monitor is sealed together with the water sample into a container. The air filled radon monitor is protected from the water by a thin radon permeable rubber foil. The dissolved radon and

the radium content of water samples is determined by two subsequent exposures. In the first exposure the sum of dissolved radon and radium concentrations, in the second exposure the radium concentration itself are measured. I have developed a model, which describes the temporal variation of the radon concentration in the measuring volume of the small radon monitor. For the given arrangement at the Radiochemical Department of Veszprém University I have determined experimentally the calibration coefficient, 24.1 $tracks \cdot cm^{-2}/(d \cdot kBq \cdot m^{-3})$ [A15], which value coincided well with the calculated one. Characteristic for the method is that it not applies any type of separation procedures, the use of which is often difficult in field circumstances.

5 Tézisek

(Az [A...]-ként hivatkozott új tudományos eredményeket tartalmazó cikkek másolatai a mellékletben találhatók.)

1. A karsztbarlangok légterében mért radonkoncentráció értékek jellemzése

1.a Áttekintettem az elmúlt két évtizedben publikált barlangi radonmérési programok eredményeit s megállapitottam, hogy a világ 220 barlangjában mért radonkocentráció adatok eloszlása lognormális [A1, A2] (GM=1130 $\mathrm{Bq \cdot m^{-3}}$, GSD=6,3). Az eloszlás típusa megegyezik más geokémiai elemek szokásos földi eloszlásával. A skála alsó részét nagy térfogatú termek ill. erősen szellőző helyek, míg a felső részt zárt, rosszul szellőző helyek és uránium gazdag mineralizációk jellemzik.

1.b Nyomdetektoros mérések alapján megállapitottam, hogy a 31 vizsgált magyarországi barlangban az éves átlagos radon aktivitáskoncentráció a 0,3-20 $\mathrm{kBq \cdot m^{-3}}$ tartományba esik, a mért radon idősorok periodicitása tipikusan egy vagy fél év. Jellemző az éven belüli 2-50 radonkoncentráció maximum/minimum arány [A3]. Az barlangok többségében nyári maximumot és téli minimumot tapasztaltam, ritkábban fordítva.

1.c Minden egyes vizsgált barlangban a mért évi átlagos radon aktivitáskoncentráció hosszú idejű változását tapasztaltam [A1, A4]. Ez a hosszúidejü trend eltérő lehet egy adott barlang különböző pontjain. Az utóbbi jelenséget jól mutatja a Sátorkő-puszta-barlang két különböző mélységében felvett radon idősor (lásd 4. ábra [A1]). A két adatsornak az adatpárok átlagára normált különbsége időben növekvő tendenciát mutat. A megfigyelt jelenség tükrözi a lassan változó környezeti paraméterek hatását a radontranszport folyamatokra. Ilyen paraméter lehet a kőzetek radonkibocsátási tényezőjének a lassú változása a barlangot befoglaló kőzet víztartalmának változása miatt. Ez utóbbi paraméter értéke a klimatikus változások miatti éves csapadékmennyiség változását tükrözheti.

2. A karsztosodás fokának és a barlang morfológiájának hatása a barlangi levegő radonkoncentrációjára

2.a Modellszámításokkal kimutattam, hogy a repedésekben kialakuló telítési radonszintek és a radon exhalációja a repedésekből erősen függ a repedés méretétől. Kis repedésméret/légáramlási sebesség arányoknál, a repedés menti advektív radontranszportot erősen csökkenti a keresztirányú diffúzió hatása [A1, A6] A számítások következtetéseit a Vass Imre- és a Lamalou-barlangokban folytatott radonkoncentráció mérések eredményeivel igazoltam. A Vass Imre-barlang egy gyengén fejlett repedésrendszerrel jellemezhető, a Lamalou karszt, Franciaország, pedig egy erősen karsztosodott terület nagy oldási üregekkel és töredezett rétegekkel. A Vass Imre barlangban felvett radon idősorokra a nagyfokú napi stabilitás a jellemző a megfigyelt erős napi légáramlás változások ellenére. A jelenség a diffúzió erős simító hatását mutatja. Ezzel szemben a Lamalou barlangban, a felszíni hőmérséklet napi változásainak megfelelően, erős napi radonszint változásokat találtunk [A7].

2.b Vízszintes barlangokban a napi átlagos radonkoncentráció ugrásszerűen változik a napi átlagos felszíni hőmérséklet függvényében, egy átmeneti tartománnyal az alacsony téli és magas nyári értékek között. Mérésekkel rámutattam, hogy az átmeneti tartomány szélessége függ a felszínnel kapcsolatban álló függőleges repedésrendszerek számától [A8]. A Vass Imre-barlangban ennek az átmeneti tartománynak a szélessége 5 °C, az Abaligeti-barlangban pedig 10 °C. A Vass Imre-barlangban csak egy, a barlang vége felé elhelyezkedő repedésrendszer található. Ezzel szemben az Abaligeti-barlangban több mint egy, jobban fejlett függőleges törészóna található. Az átmeneti tartomány szélesedését a repedések számának és méretének függvényében a Szemlőhegyi-barlangban mért adatok is alátámasztják. Itt a megfigyelt görbe gyakorlatilag lineáris (lásd 3. ábra [A8]).

3. Légáramlási utak és mikroklimatikus zónák azonosítása barlangrendszerekben radonkoncentráció mérések alapján

3.a Rámutattam, hogy az évszakosan váltakozó irányú légáramlás következménye a karsztos felszíneken az évszakosan változó radonexhaláció [A3]. Erre a jelenségre a legmeggyőzőbb kísérleti bizonyítékot a Hajnóczy-barlangban és a barlang feletti felszínen egy repedésben mért radonszintek ellenütemű változása szolgáltatta (lásd 6. ábra [A2]). A felszínen magas téli radonkoncentrációt mértünk, de téli maximumokat találtunk Magyarország számos más karsztos területén is. A fenti jelenség magyarázatul szolgálhat az irodalomban publikált további megfigyelésekre is.

3.b Mikroklimatikus zónák jelenlétét mutattam ki a radon idősorok analízise alapján. A Baradla-barlangban ezek a zónák [A5]: 1. az aggteleki bejárati szakasz magas nyári és alacsony téli radonkoncentráció értékekkel; 2. A Libanon-hegy és a vöröstói bejárat közti rész időben stabil radonszintekkel; valamint a 3. jósvafői és vöröstói bejáratok közötti rész magas téli és alacsony nyári radonszintekkel. A radonkoncentráció mérések alapján rámutattam, hogy légkörzés szempontjából kapcsolat van a Baradla barlang és az alatta elhelyezkedő Rövid-Alsó-barlang között. Mikroklimatikus zónák jelenlétét a tavaszi radonkoncentráció értékek szórásának különbözősége alapján sikerült kimutatnom a Létrási-Vizes-barlangban is [A2, A9].

4. A felszín alatti légáramlási sebesség meghatározása és kapcsolata az éves átlagos radonkoncentrációval

4.a Meghatároztam a friss külszíni levegő barlangba való behatolásának terjedési sebességét és egy vertikális barlang térfogatát a folyamatos radonkoncentráció mérések alapján. Ez a jelenség vízszintes barlangokban a külső hőmérsékletnek a barlangi hőmérséklet alá való csökkenésekor, függőleges barlangokban az atmoszférikus légnyomás meredek növekedésekor tapasztalható. A detektorok közötti távolságot és a radonszint esések közötti időeltolódását figyelembe véve a légáramlás sebessége a Vass Imre barlang főjárata mentén kb. 50 m·h^{-1} [A1, A2]. Mivel a járat hossza 600 m, ezért külszíni hatások 12 óra alatt érik el a barlang végpontját. Ezzel szemben a Cserszegtomaji-kútbarlangban a külszíni hatások terjedési sebessége a kút aljától 10-20 m-re csak kb. 2 m·h^{-1}. Ez az alacsony érték mutatja, hogy a barlang szellőzése rossz. A mérések szerint átlagosan 2 hPa légnyomás emelkedés hatására terjed ki a friss levegő radonkoncentráció hígító hatása a bejárati kút teljes térfogatára. Az ideális gáztörvény figyelembevételével a barlang térfogata eszerint kb. 10000 m^3, ami egy nagyságrenddel nagyobb, mint az ismert barlangi járatok térfogatának becsléséből számított érték.

4.b Kimutattam, hogy kapcsolat van a felszín alatti évi átlagos radonkoncentráció és felszín alatti légáramlások erőssége között [A2, A8]. A Vass Imre-barlangon átáramló levegő mennyiségét a barlangban található szifon természetes módon szabályozza. Ha a szifon zárva van, nincs mérhető légáramlás a barlangban. A légáramlás természetes korlátozása éves szinten mintegy 30%-os átlagos radonkoncentráció csökkenést okoz, egyben csökkentve az évszakos radonkoncentráció változások nagyságát. Ez a jelenség a fedő rétegeken keresztüli légkörzéssel magyarázható s a periodikusan változó áramlási feltételek átlagos radonkoncentrációt növelő hatását mutatja. A legmélyebb magyarországi függőleges barlangok (Szepessy, Istvánlápa) mélyén talált alacsony radonkoncentráció szintek és a radonkoncentráció szezonális változásainak a hiánya [A3] szintén a fenti jelenségnek tulajdonítható, mivel szellőzés szempontjából a mély barlangok általában a legzártabbak közé tartoznak.

5. Radontranszport vizekben

5.a Kimutattam, hogy a szellőzési folyamatokon túl, esetenként, a barlangi vízfolyások is befolyásolják a barlangi levegő radonszintjét [A1, A2, A4, A9]. A Létrási-Vizes-barlang mélyebb részein a radonszintek évszakos periodikus változása jelentősen módosul, a barlang végpontja felé a periodikus jellege csökkenése mellett nő a tavaszi értékek szórása. Ezzel szemben nagy hasonlóság van a barlang végpontján fakadó egyik időszakos forrás vízhozama és a levegőben mérhető radonszint időbeli változásai között. Mivel a felszín alatti vizek oldott radontartalma jelentősen nőhet a porózus kőzeteken való áthatolásuk során, így a hasonlóság azzal magyarázható, hogy a radont ez az időszakos patak szállítja a helyszínre.

5.b Egy 270 m mély fúrt karsztkútban folytatott radonkoncentráció profil mérése alapján keveredési folyamatokat azonosítottam a függőleges vízoszlopban [A4, A10]. Az adatok értelmezéséhez kifejlesztettem egy eljárást, amely kapcsolatot teremt a mért radonkoncentráció profil és a mélységfüggő vertikális transzportsebesség között. Az eredmény a radonnak a függőleges vízoszlop menti 0,7 m·h^{-1} átlagos sebességű felfelé irányuló mozgását jelzi. Megállapítottam, hogy a megfigyelt nagy vertikális sebesség a vertikális termikus gradiensek indukálta konvekciónak tulajdonítható, a geogáz mikrobuborékok által gyorsított radon feláramlás lehetőségét elvetettem. A vertikális keveredés erős hatását egy 8,5 m magas modelltornyon (Montpellieri Egyetem, Franciaország) lefolytatott kísérletsorozat is igazolta [A11]. Mivel a 80-as években Magyarországon az urán kutatás részben a sekélyfúrásokban felvett víz alatti radonkoncentráció profilok értelmezésén alapult, az utóbbi eredmény felhasználást nyert a módszer ipari alkalmazásában.

6. Módszerfejlesztések

6.a Jelentősen hozzájárultam a szilícium fotodiódán alapuló Dataqua radonmérő eszköz kifejlesztéséhez [A12]. A fotodióda alkalmazhatóságának ötletét terepi radonkoncentráció mérésekre (alfa részecskék detektálása útján) a francia együttműködő partnereinktől (Montpellieri Egyetem, Hidrogeológiai Tanszék, Montpellier, Franciaország) kaptuk. A műszer hardwerét és szoftverét a Dataqua Elektronikai Kft. fejlesztette ki. A mérőfej érzékeny térfogatának tervezését modellszámításokkal, az elkészült mérőfejet a mérőtérfogat és az elektronikus beállítások változtatásával kísérletileg teszteltem. A Svéd Nemzeti Sugárvédelmi Intézet stockholmi referencia radonkamrában folytatott méréseim alapján meghatároztam a műszer kalibrációs állandóját, 6,7 cph/kBq·m^{-3}·h, mely jó egyezésben volt a számításaimmal. A Dataqua márkanevű műszer prototípusát terepi körülmények között a Mecsekurán kft. (Pécs) munkatársa, Dr. Várhegyi András tesztelte.

6.b. Jelentősen hozzájárultam a radon vékony fóliákon belüli diffúziós állandójának nyomdetektor technikán alapuló mérését szolgáló új eljárás kifejlesztéséhez [A13]. A mérés során a vizsgálandó fóliát egy radonforrás és egy mérőkamra közé helyezzük. Kidolgoztam a módszer hátterét képező modellt, amely leírja a radonkoncentráció időbeli változását a mérőkamrában a mérendő diffúziós állandó függvényében. Az eljárás lehetővé teszi a vizsgálati idők csökkentését néhány órára. A módszer alkalmazhatóságát különböző vastagságú polietilén fóliákkal laboratóriumi körülmények között teszteltem. A kapott érték, 7,1*10^{-8} cm^{2}·s^{-1}, jól egyezik az irodalomban publikáltakkal. A különböző anyagok radonáteresztő képessége a radont a lakásokból kizáró szigetelő anyagok iránti igény miatt érdekes. A módszer gyakorlati alkalmazásaként megvizsgáltuk néhány padlóburkoló műanyag radonáteresztő képességét. Úgy találtuk, hogy mindegyik anyag 2-3 nagyságrenddel csökkenheti a talajeredetű forráshoz köthető radonszintet a lakásokban [A13].

6.c Jelentősen hozzájárultam a radon polimer/szilikát gélekben és agyag szuszpenziókban való effektív diffúziós állandójának mérését szolgáló két további eljárás kifejlesztéséhez [A16, A17]. Hasonlóan az előbbi eljáráshoz, a vizsgálandó anyagot egy radonforrás és egy mérőkamra közé helyeztük. Nyomdetektorokkal mértük a radonexpozíciót a minták két oldalán. A mintán belüli diffúziós tömegtranszportot numerikus modelleztük különböző mintavastagság és besugárzási idők esetére. A diffúziós állandót, mint paramétert, a mért expozíciókhoz tartozó legjobban illeszkedő modellezés eredménye szolgáltatta. A kísérleti elrendezés zártságát Dataqua típusú folyamatos radonmérőkkel felvett idősorok elemzése alapján ellenőriztem. Az elemzés a besugárzási idő rész-időintervallumaira meghatározott diffúziós paraméter állandóságának vizsgálatán alapult. Úgy találtuk, hogy a vizsgált polimer/szilikát gélt tartalmazó porózus közegben az effektív diffúziós állandó 3,3*10^{-6} cm^{2}·s^{-1}, míg a Montax/agyag szuszpenzióban 6,0*10^{-6} cm^{2}·s^{-1}. Az eredmények alapján a tiszta fázishoz tartozó értékek összemérhetők a pusztán vizes oldatokat jellemző adattal, ezért az ezeket az anyagot alkalmazó szigetelési eljárások nagyon vonzóak lehetnek.

6.d. Jelentősen hozzájárultam a vízminták rádium és oldott radon tartalmának mérését szolgáló, nyomdetektor technikán alapuló új módszer kidolgozásához [A14]. A mérés során a vízmintát egy kis térfogatú, gumimembránba csomagolt radon monitorral együtt egy üvegedénybe zárjuk. A gumimembrán átengedi a radont de kizárja a vizet a radonmonitor mérőtérfogatából. A vízminta oldott radon és rádium tartalmát két független méréssel határozzuk meg. Az első méréssel a rádium és oldott radon együttes koncentrációját, a második méréssel a rádium koncentrációt határozzuk meg. Kidolgoztam a kis térfogatú radon monitor mérőtérfogatán belüli a radonkoncentráció időbeli változását leíró modellt. Az általunk használt mérési elrendezésre a Veszprémi Egyetem Radiokémiai Tanszékén folytatott mérésekkel meghatároztam modellben szereplő kalibrációs állandó értékét, 24,1 nyom·cm^{-2}/nap·kBq·m^{-3} [A15], mely jó egyezésben volt számításaimmal. A kidolgozott módszerre jellemző, hogy semmilyen elválasztási technikát nem alkalmaz, melyek használata terepi körülmények között gyakran nehézkes.

6 References

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RARE GASES GEOCHEMISTRY

IV INTERNATIONAL CONFERENCE

8 - 10 October, 1997

University of Roma TRE
Rome, Italy.

General Information

Programme Overview

Book of Abstracts

PROGRAMME

TUESDAY 7 OCT.

REGISTRATION
17.00 - 19.00

WEDNESDAY 8 OCT.

REGISTRATION
08.00 - 09.30

OPENING CEREMONY
09.30 - 10.00

COFFEE BREAK
10.00 - 10.30

ORAL PRESENTATIONS
10.30 - 12.30

LUNCH
12.30 - 14.00

ORAL PRESENTATIONS
14.00 - 16.00

COFFEE BREAK
16.00 - 16.30

ORAL PRESENTATIONS
16.30 - 18.30

THURSDAY 9 OCT.

ORAL PRESENTATIONS
08.45 - 10.30

COFFEE BREAK
10.30 - 11.00

ORAL PRESENTATIONS
11.00 - 12.30

LUNCH
12.30 - 14.00

ORAL PRESENTATIONS
14.00 - 16.00

COFFEE BREAK
16.00 - 16.30

POSTER PRESENTATIONS
16.30 - 18.30

INTERNATIONAL COMMITTEE MEETING
18.30 - 19.00

FRIDAY 10 OCT.

ORAL PRESENTATIONS
08.45 - 10.30

COFFEE BREAK
10.30 - 11.00

ORAL PRESENTATIONS
11.00 - 12.45

LUNCH
12.45 - 14.00

ORAL PRESENTATIONS
14.00 - 15.30

COFFEE BREAK
15.30 - 16.00

ORAL PRESENTATIONS
16.00 - 17.30

CONFERENCE BANQUET
19.45 - 23.30

SATURDAY 11 OCT.

EXCURSION TO THE UNDERGROUND LABORATORY OF GRAN SASSO
08.30 - 16.30

Conveners:

Francesco **Bella** - Physics Department - University of Roma TRE - Italy.
Giorgio **Ferrara** - Earth Science Department - University of Pisa - Italy.
- Institute of Geochronology and Isotopic Geochemistry, CNR, Italy.

General Chairman:

Giovanni **Martinelli** - Regione Emilia-Romagna, Bologna - Italy.

International Committee:

G. Akerblom (Sweden), P. Allard (France), I. Barnet (Czech Rep.), A. A. Belyaev (Russia), P.F. Biagi (Italy), A. Chambaudet (France), M. Charlet (Belgium), M. Delcourt-Honorez (Belgium), J. Heinicke (Germany), I. Hunyadi (Hungary), R. Ilic (Slovenia), C. Y. King (USA), J. Lebecka (Poland), G. Martinelli (Italy), J. Matsuda (Japan), J.Miles (U.K.), M. Monnin (France), C. Papastefanou (Greece), N. Segovia (Mexico), H. Surbeck (Switzerland), L. Tommasino (Italy), T. Torgersen (USA), H. S. Virk (India), H. Wakita (Japan), W. Zhang (China).

Local Organizing Committee:

F. Bella (University of Roma TRE), P. F. Biagi (University of Bari), A. Ermini (University of Roma-Tor Vergata), A. Esposito (INFN-LNF), G. Ferrara (University of Pisa), G. Martinelli (Regione Emilia-Romagna), L. Tommasino (ANPA).

Scientific topics

Session I: **Rare gases related to earthquakes, volcanoes and environment.**
Chairman: Pier Francesco **Biagi**
Session II: **Radon in Radioprotection.**
Chairman: Luigi **Tommasino**
Session III: **Rare gases in mantle fluids.**
Chairman: Giovanni **Martinelli**
Session IV: **Measurement techniques and instrumentation.**
Chairman: Adolfo **Esposito**

X→

FRIDAY 10 OCTOBER 1997

LECTURE ROOM

Chairperson: *J. Heinicke*

08.45 **Hydrogeological investigations of the German earthquake task force after the October 1, 1995 Dinar earthquake, SW Turkey.**
O46 H. Woith, C. Milkereit, U. Maiwald, A. Pekdeger
09.00 **Radon measurements in association with earthquakes.**
O47 C. Papastefanou, M. Manolopoulou, S. Stoulos, A. Ioannidou, E. Gerasopoulos
09.15 **Groundwater Helium content related to the Spitak (Armenia) and Karymsky (Russia) earthquakes.**
O48 P.F. Biagi, F. Bella, E. Cozzi, A. Ermini, G. Martinelli, Y.M. Khatkevich, E.I. Gordeev, D. Zilpimiani
09.30 **Analysis of Argon concentration anomalies in underground water in Kamchatka (Russia).**
O49 S.P. Kingsley, C.W. Anderson, P.F. Biagi, P.J. Derlien, A. Ermini, E.I. Gordeev, Y.M. Khatkevich
09.45 **Radon measurements in soil and water in the seismic Friuli area.**
O50 M. Garavaglia, C. Braitenberg, M. Zadro, B. Porfidia, F. Quattrocchi, M. Calcara
10.00 **Earthquake related Radon monitoring in the Dead Sea area.**
O51 U. Vulkan, B. Lang, G. Steinitz
~~10.15 **Enhancement of Radon and aerosol degasations along a gesture can be used in prediction of earthquake and of volcanic eruption.**~~
~~**O52** V.A. Alekseev, N.G. Alekseeva~~

COFFEE BREAK

Chairperson: *P.F. Biagi*

11.00 **Ground fluid precursors before the 1995 Menglian, Yunnan, earthquake (M= 7.3).**
O53 R. Lin, C. Lide ([illegible])
11.15 **Radon/Helium studies for earthquake prediction in N-W Himalaya.**
O54 H.S. Virk
11.30 **The precursor effects in the mineral spring "Radonquelle", Bad Brambach prior to the January 14-19, 1997 earthquake swarm near Novy Kostel (NW Bohemia).**
O55 U. Koch, J. Heinicke
11.45 **Regular oscillations of helium content of ground water as a new geochemical precursor of an earthquake.**
O56 A. Belyaev
12.00 **Research on the relationship between escaping Radon and crustal stress-strain.**
O57 W. Zhang
12.15 **The correlation between variation of Radon content in groundwater and earthquakes and its indefiniteness.**
O58 Z. Zhang

LUNCH

Chairperson: *N. Segovia*

14.00 **Thermal effects on Radon transport in soil.**
O59 I. Csige, J. Hakl, A. Vasarhelyi, I. Hunyadi, J.L. Seidel, H. Climent, M. Monnin, A. Varhegyi
14.15 **The sealing of soils and its effect on Radon migration in soil.**
O60 J. Wiegand
14.30 **Characteristic of near-surface Radon transport in temperate zone under continental climate.**
O61 I. Hunyadi, I. Csige, G. Geczy, J. Hakl, L. Lenart, A. Vasarhelyi
14.45 **Comparative study of indoor Radon concentrations in four Radon prone areas in Central Europe.**
O62 I. Hunyadi, I. Csige, Z. Dezso, J. Hakl, L. Lenart, I. Mocsy, Z. Nemeth, Z. Papp, J. Somlai, A. Vasarhelyi
15.00 **One year of Radon measurements indoor and outdoor in Lombardy.**
O63 L. Sesana, U. Facchini
15.15 **Study on Rn-222 density in the soil.**
O64 L. Sesana, L. Boschiroli, R. Valsecchi, U. Facchini

COFFEE BREAK

Chairperson: *I. Barnet*

16.00 **Rn222 emissions from mud volcanoes in northern Apennines: a time series analysis.**
O65 V. Lapenna, D. Albarello, G. Martinelli.
16.15 **Seasonal Radon concentration changes in Niedzwiedzia Cave (SW Poland).**
O66 T.A. Przylibski, W. Ciezkowski
16.30 **Site specific Radon regimes of cave systems.**
O67 J. Hakl, I. Csige, A. Varhegyi, I. Hunyadi, Zs. Kertesz, G. Geczy
16.45 **Origin of radiogenic Helium in deep aquifers and the problem of dating very old ground water.**
O68 D. Pinti, B. Marty
17.00 **Seismic waves in the urban environment triggering Radon release from the soil.**
O69 S. Schmid, J. Wiegand
17.15 **Theoretical dependencies of response function Vt/Vo on additional gamma-exposure and etching temperature for polyethyleneterephthalate detector.**
O70 V.A. Ditlov

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SITE SPECIFIC RADON REGIMES OF CAVE SYSTEMS

J. Hakl, I. Csige, A. Várhegyi[*], I. Hunyadi, Zs. Kertész and G. Géczy[•]

Institute of Nuclear Research, H-4001 Debrecen, POB. 51, Hungary
[*]Mecsekurán Ore Mining Ltd., H-7614 Pécs, POB. 65, Hungary
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H-1083 Budapest, Ludovika Square 2, Hungary

We have measured spatial and temporal variations of ^{222}Rn concentration in eight, morphologically and genetically different caves in Hungary. Our results show, that the influence of meteorological parameters on radon in caves largely depends on cave morphology, for horizontal caves surface temperature, for vertical caves barometric pressure variation controlled driving forces are dominant. Furthermore, at different parts of a particular cave system there are differences in the way how radon concentration reacts to changes in control parameters. Based on the patterns of temporal and spatial changes, distinct microclimatic zones can be delineated, which are characterized by site specific radon regimes. It was observed, that the long term stability of such a microclimatic zones can be influenced by changes in the penetrability of the underground void system. Our measurements further show, that similarities in radon regimes of two, separate caves may indicate possible connection between the two systems.

This work was supported by the Hungarian National Scientific Research Fund contracts Nos. T 016558 and T 017560

ORIGIN OF RADIOGENIC HELIUM IN DEEP AQUIFERS AND THE PROBLEM OF DATING VERY OLD GROUND WATER

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[°°] Centre des Recherches Pétrografiques et Géochimiques, Vandoeuvre-Les-Nancy, France
Ecole Normale Supérieure de Géologie, Nancy Cedex, France

Noble gases, inert elements having isotopes produced by decay of long-lived radionuclides, offer a unique tool for tracing the fluid circulation in deep aquifers and dating very old ground water in the range of 104-108 a. Several works have shown that helium water age, calculated from the accumulation rate in water of radiogenic 4He produced in the aquifer rocks, is frequently higher than the hydrologic age. This discrepancy is generally interpreted following two contrasted models: i) heterogeneities of aquifers, which allow water stagnation and accumulation of large amounts of radiogenic 4He or ii) addition of exotic helium from deeper regions such as the continental crust. Resolving ultimately the sources of radiogenic 4He in sedimentary fluids is fundamental for the correct calibration of this geochronometer in basin hydrology.

In this contribution, we propose that the apparent contrast between He ages and hydrologic ages reflect mixing of different types of waters, having different residence times. In the Paris Basin, northern France, we show, using the helium isotope ratios, that such a mixing occurs between two different aquifers having constrasted helium contents, each of them being heterogeneous in term of chemical composition and permeabilities. Differences in the radiogenic isotopic 4He/40Ar* ratios between these two aquifers strongly suggest that a significant fraction of 4He is produced internally in the aquifer rocks, and imply residence times for ground water much longer (1-50 Ma) than those obtained from hydrologic studies (1Ka - 1Ma). Independent fluid age estimates, based on the water paleotemperatures at the recharge calculated with the atmosphere-derived noble gases, seems confirm the presence of very old ground waters in the Paris Basin.

Rare Gas Geochemistry IV International Conference, 8-10 October, 1997
University of Roma TRE
Rome, Italy

SITE SPECIFIC RADON REGIMES OF CAVE SYSTEMS

J. Hakl, I. Csige, A. Várhegyi[1], I. Hunyadi, Zs. Kertész
and G. Géczy[2]

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1 - Mecsek Ore Mining Ltd, Pécs, Hungary
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Budapest, Hungary

Sopported by: Hungarian National Scientific Research
Fund contract Nos. T 016558, T 017560

- microclimatic zone : given temporal pattern of external parameters (air flow ⇒ T_{cave}, humidity, Rn ...)
- spatial stability ?

(changes in radon concentration ***regimes*** could be explained by variations in cave climate)

Vass Imre cave - Aggtelek Karst, Hungary (horizontal, dominated by chimney effect winds)

typical temporal pattern: annual change with summer maxima and winter minima

⇔ identify air flow direction

spatial pattern ⇒ winter : entrance and 'cave' zone

siphon: closed ⇔ open : is the border unchanged ?

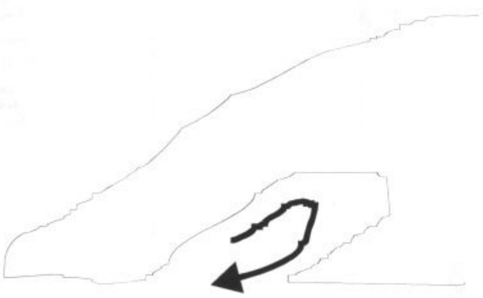
! Rn amplitude change ⇔ air flow strenght change

⇓

the change in the penetrability of the cave system may affect the microclimatic zonation of the cave⇒ as air flow ⇒ penetrability of external (polluting) effects is changed

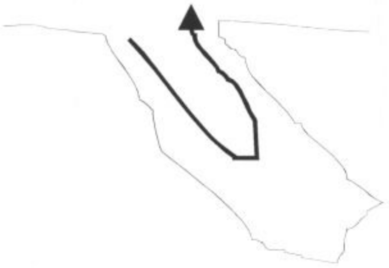
Cave Morphology

Blind end; T,p(<)

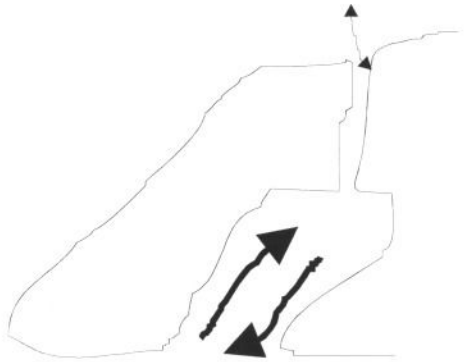


s < w

s > w

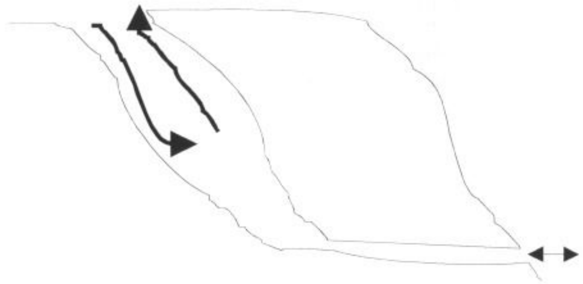


Flow through, T

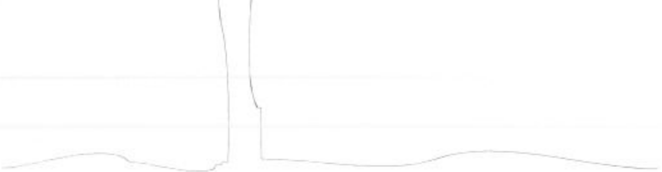


s > w

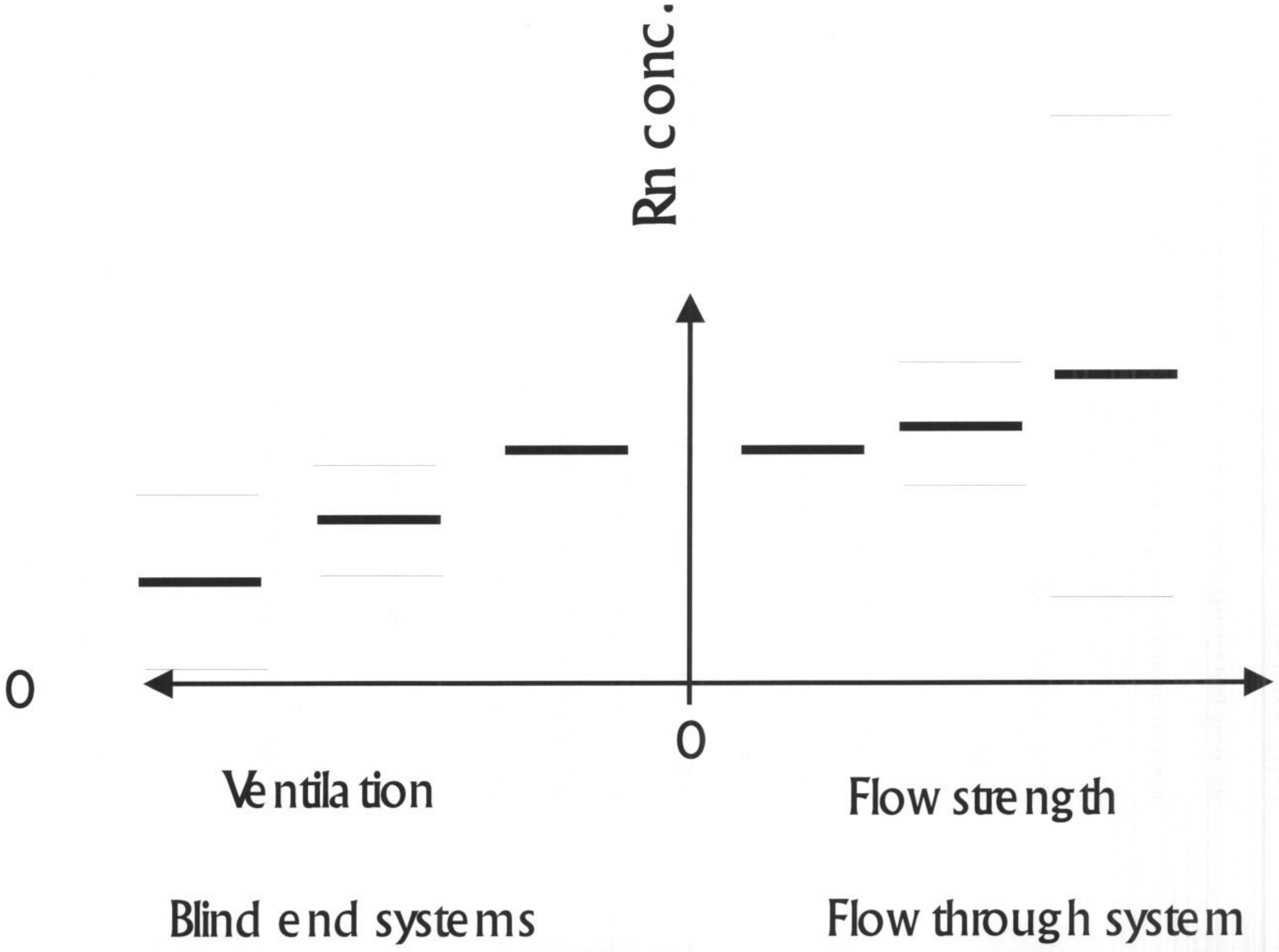
s < w



P dominates



Ventillation effect



! behind siphon phenomenon in winter 1989/90:

NO MINIMUM ⇒ possibility of change in the microclimatic zonation

Baradla cave - Aggtelek Karst, Hungary

annual average: 1 - 5,3 $kBqm^{-3}$

(min) three distinct microclimatic zones:
Aggtelek part (=Styx) : summer max, winter min
Nehéz út - Vöröstó entrance: stable levels with increasing trend
- *Retek and Törökmecset branches have no effect on the radon records* -
Vöröstó - Jósvafő entrance: summer min, winter max with increasing trend

Pálvölgyi cave
Buda Hills, Hungary
summer maxima, winter minima, monthly averages - T, p?
annual average: 0.7 - 3.1 increses with depth
annual max/min ratio: 1.4 - 15.5 varies with depth
(different patterns of the T driven process)

Mátyáshegyi cave

Buda Hills, Hungary

summer maxima, winter minima, monthly averages - T, p?

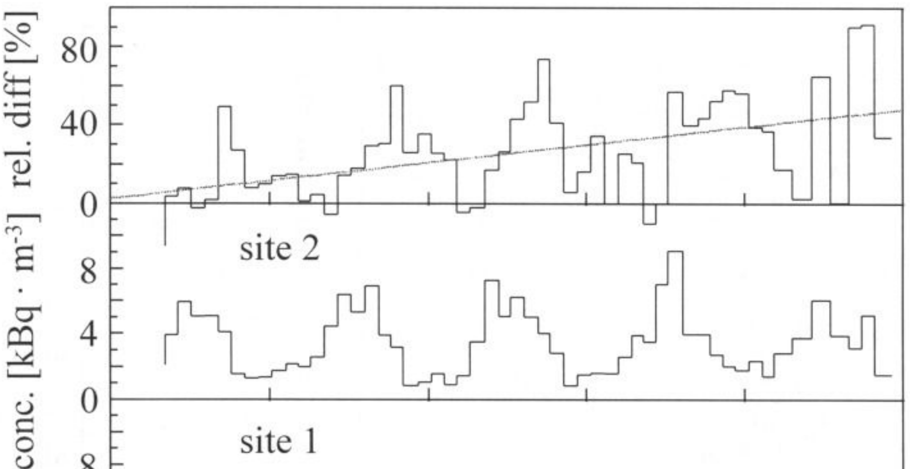
annual average: (2.3) 3.2 - 3.8 spatially stable

annual max/min ratio: 1.4 - 2.1 stable

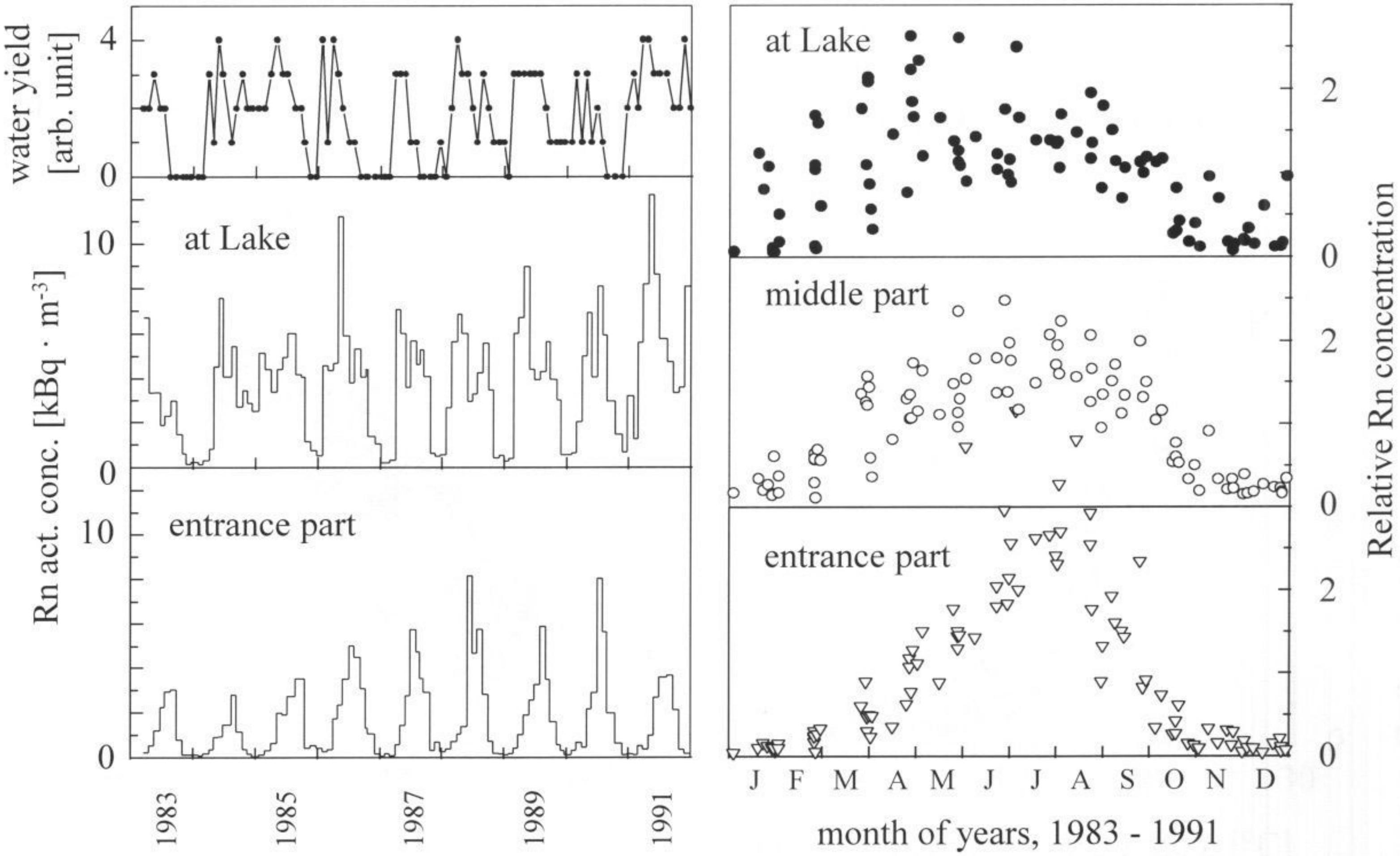
(similar patterns of the T driven process)

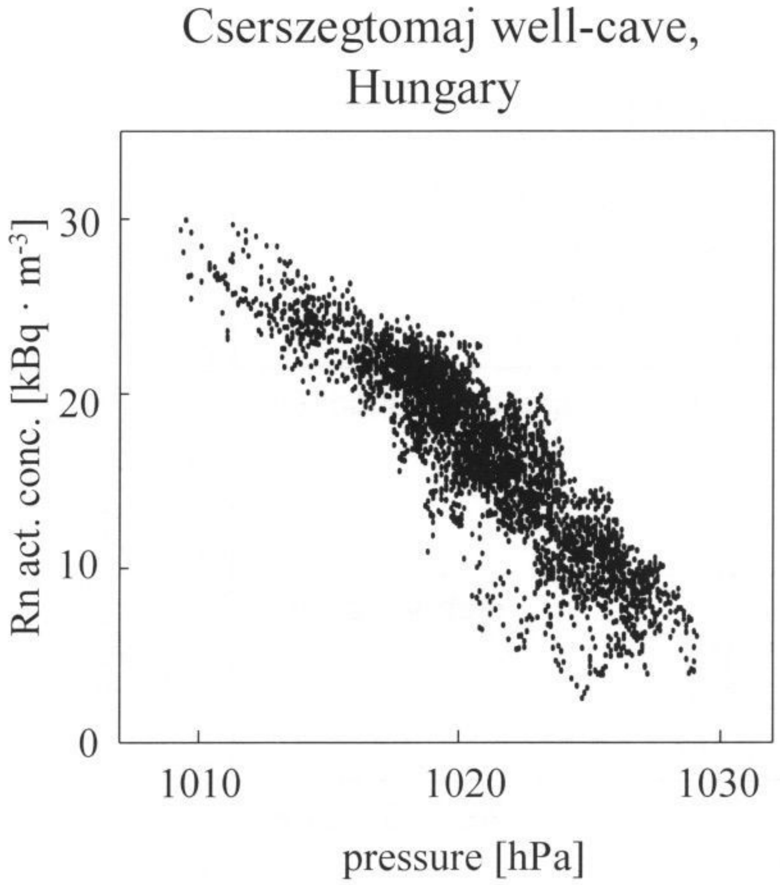
? connection between the two caves

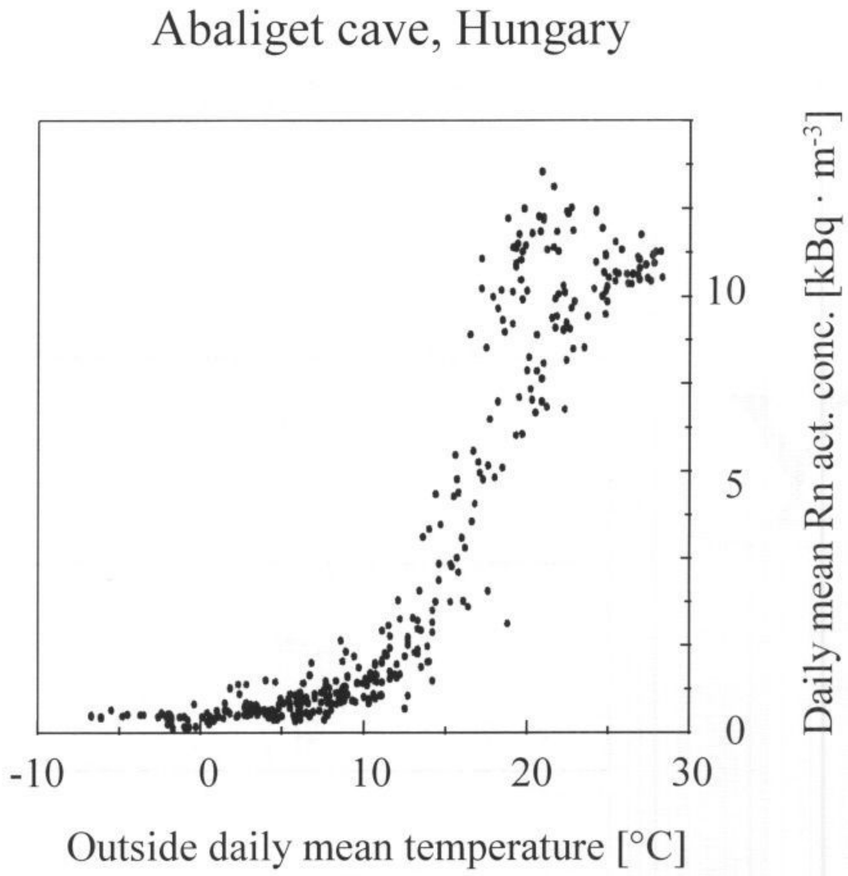
Sátorkő-puszta cave, Hungary

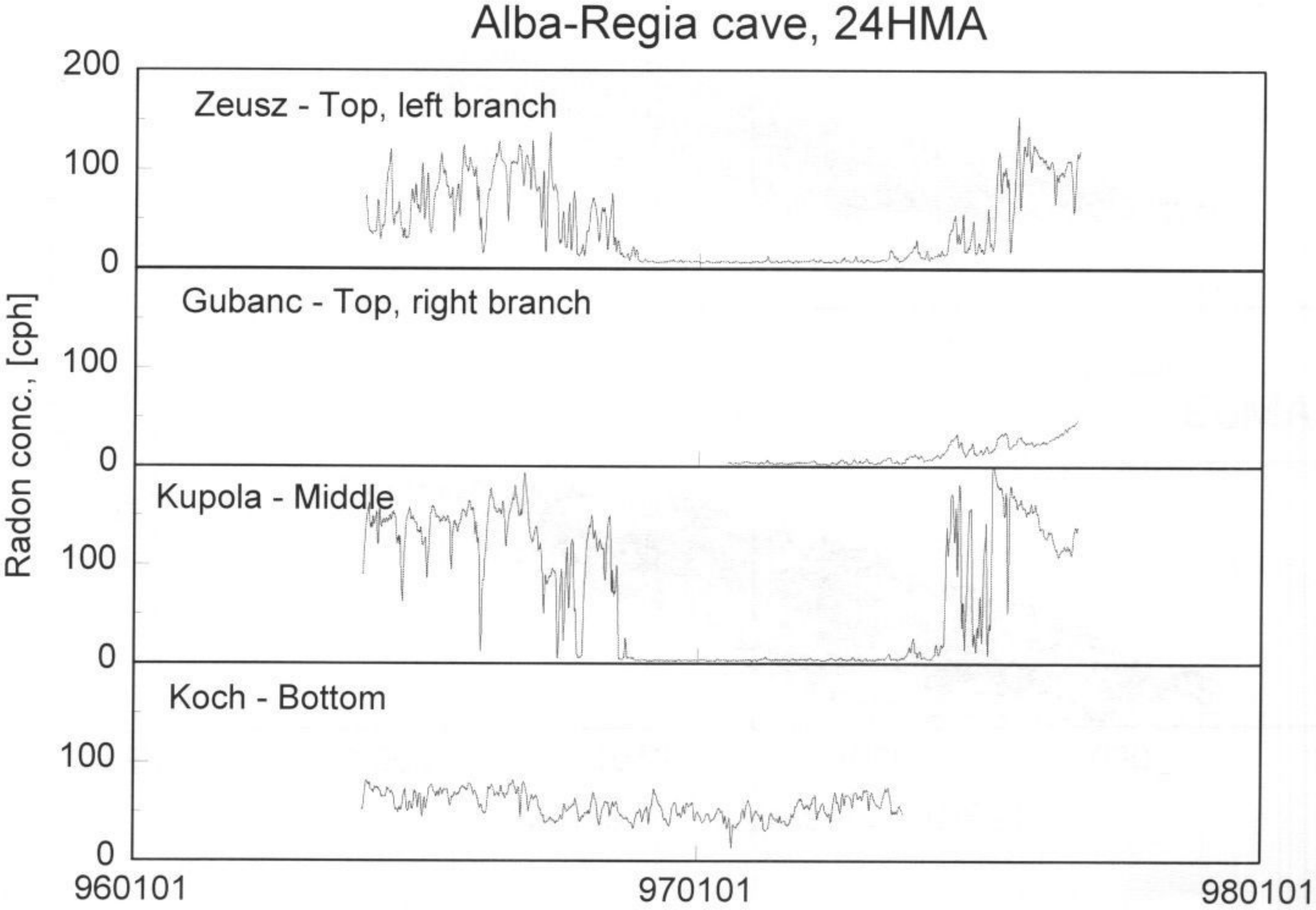


Létrási-Vizes cave, Hungary

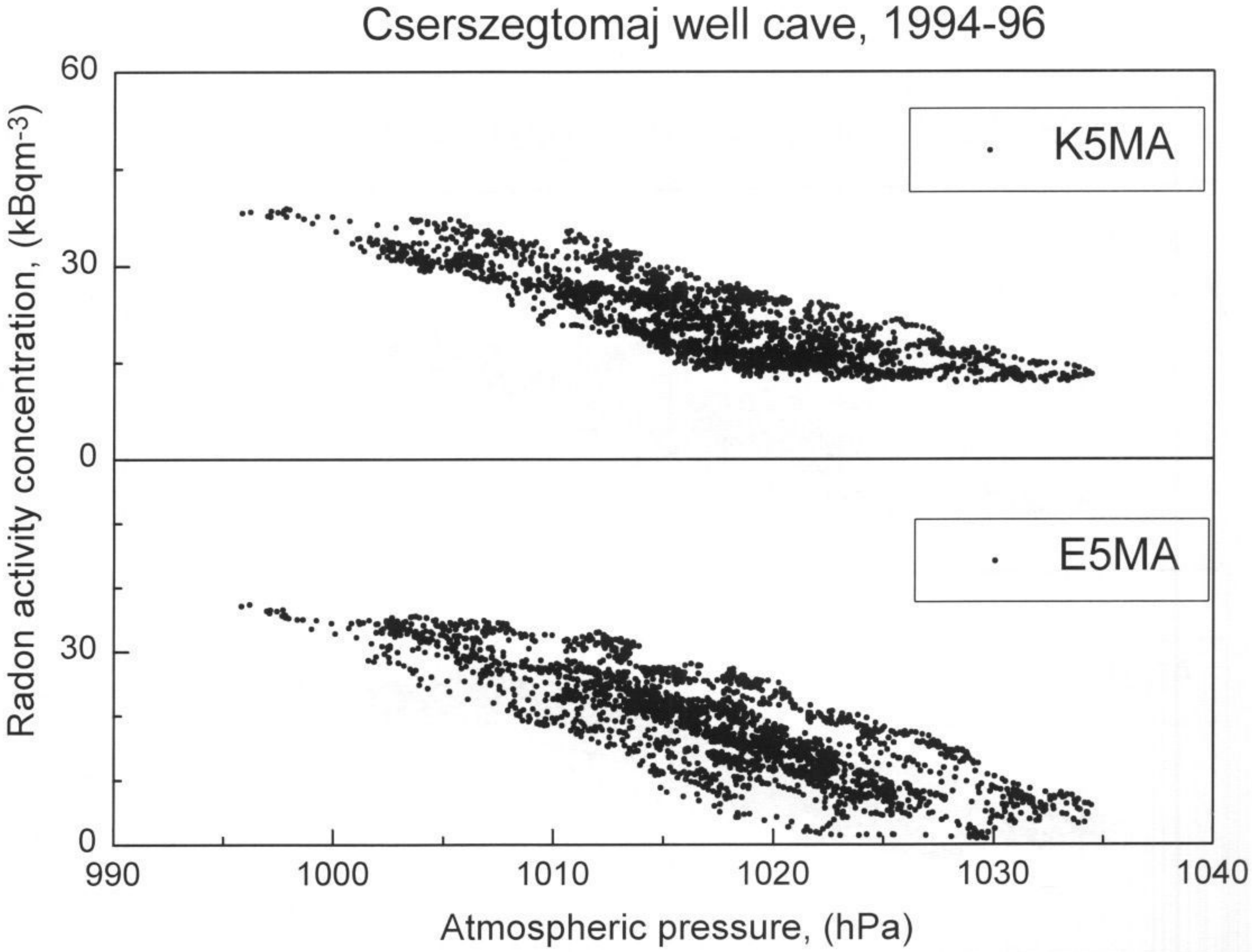


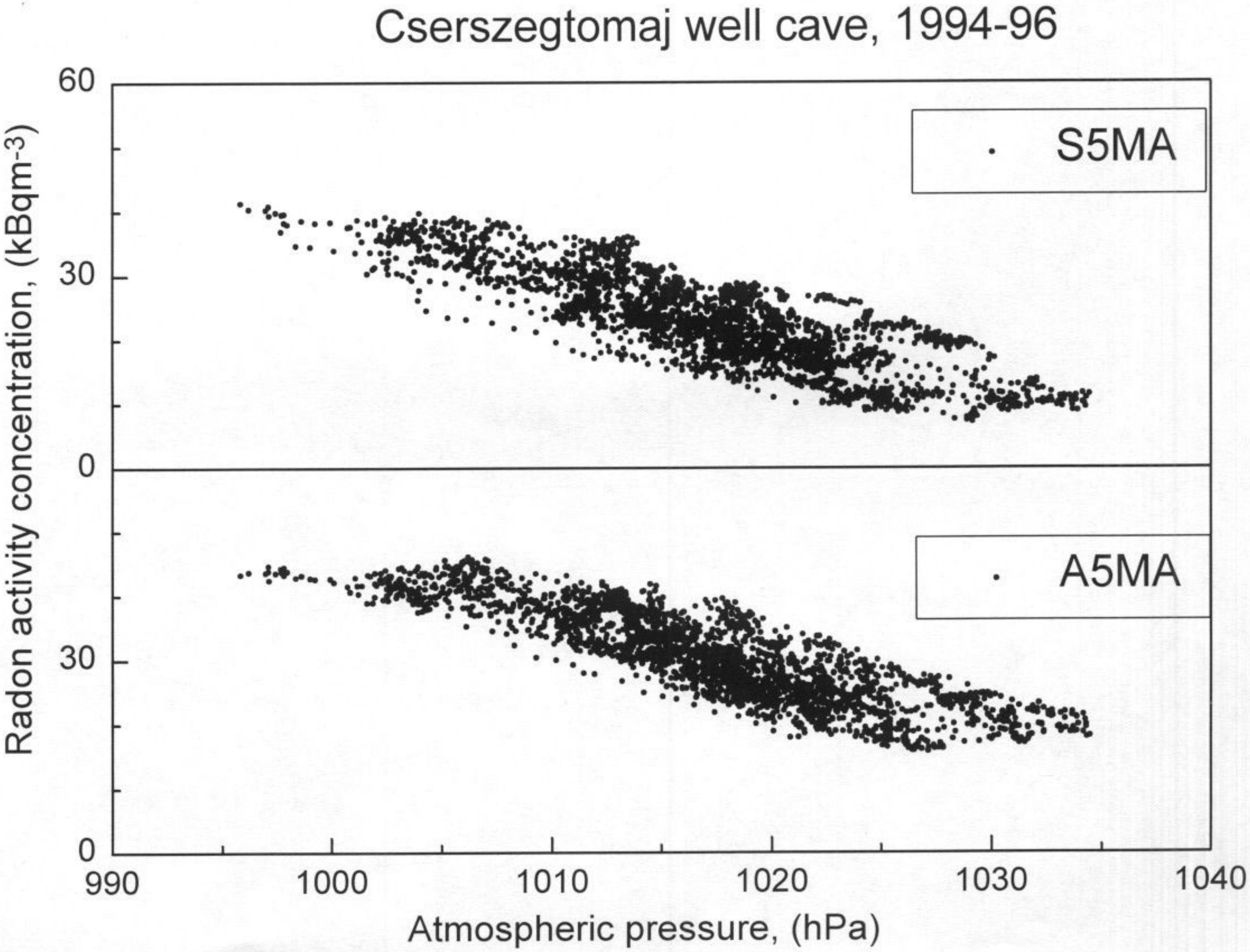

Cserszegtomaj well-cave, Hungary
Rn act. conc. [kBq · m⁻³]
30
20
10
0
1010
1020
1030
pressure [hPa]


Abaliget cave, Hungary
Daily mean Rn act. conc. [kBq · m⁻³]
10
5
0
-10
0
10
20
30
Outside daily mean temperature [°C]


Alba-Regia cave, 24HMA
Radon conc., [cph]
Zeusz - Top, left branch
Gubanc - Top, right branch
Kupola - Middle
Koch - Bottom
200
100
0
100
0
100
0
100
0
960101
970101
980101

Y


Cserszegtomaj well cave, 1994-96
K5MA
E5MA
Radon activity concentration, (kBqm-3)
60
30
0
30
0
990
1000
1010
1020
1030
1040
Atmospheric pressure, (hPa)


Cserszegtomaj well cave, 1994-96
S5MA
A5MA
Radon activity concentration, (kBqm-3)
60
30
0
30
0
990
1000
1010
1020
1030
1040
Atmospheric pressure, (hPa)